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Accident Generated Particulate Materials and Their Characteristics -- A Review of Background Information

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Pacific Northwest Laboratory
Operated by
Battelle Memorial Institute

Prepared for
U.S. Nuclear Regulatory
Commission

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Accident Generated Particulate Materials and Their Characteristics -- A Review of Background Information

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ABSTRACT

Safety assessments and environmental impact statements for nuclear fuel cycle facilities require an estimate of the amount of radioactive particulate material initially airborne (source term) during accidents. Pacific Northwest Laboratory (PNL) has surveyed the literature, gathering information on the amount and size of these particles that has been developed from limited experimental work, measurements made from operational accidents, and known aerosol behavior. Information useful for calculating both liquid and powder source terms is compiled in this report. Potential aerosol generating events discussed are spills, resuspension, aerodynamic entrainment, explosions and pressurized releases, comminution, and airborne chemical reactions. A discussion of liquid behavior in sprays, sparging, evaporation, and condensation as applied to accident situations is also included.

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SUMMARY

Pacific Northwest (PNL) has conducted a literature survey of the characterization of airborne materials potentially generated by accidents, and the results are presented here. In order to estimate the source term for accident scenarios, information was gathered to quantify 1) the amount of material initially airborne and 2) the particle size of the airborne fraction. The purpose of this review was to collect this information, which is widely scattered throughout the literature, and make it readily available to those people who require it for safety analysis reports and environmental impact statements.

A large body of information was available on particle generation and behavior, but very little could be considered directly applicable to define source terms. However, limited experimental work has been performed and fractional releases calculated, i.e., the fraction of the source powder or liquid that is initially airborne, which can be used to estimate the releases.

If such information is not available, assumptions are made so that other information can be used (with less confidence) to estimate releases. This review discusses the mechanisms for particle release as single events, such as spills. Only by first studying the small units can we develop an understanding of a complex accident. It is important to realize that many accidents will be comprised of multiple events interacting to either mitigate or magnify the total release.

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CONTENTS

ABSTRACT	iii
SUMMARY	v
1.0 INTRODUCTION	1.1
2.0 BEHAVIOR OF PREFORMED PARTICLES	2.1
2.1 SPILLS	2.1
2.1.1 Discussion	2.2
2.1.2 Summary	2.9
2.2 RESUSPENSION	2.9
2.2.1 Resuspension Factors	2.10
2.2.2 Resuspension Rates	2.11
2.2.3 Fractional Releases	2.15
2.2.4 Proposed Models	2.16
2.2.5 Particle Size	2.16
2.2.6 Summary	2.16
2.3 AERODYNAMIC ENTRAINMENT	2.17
2.3.1 Aerodynamic Entrainment of Surface Contamination	2.17
2.3.2 Aerodynamic Entrainment of Coal Dust	2.21
2.3.3 Entrainment from Spills	2.38
2.3.4 Relative Humidity Effects	2.38
2.3.5 Agglomerate Breakup	2.39
2.3.6 Motion of Sand	2.40
2.3.7 Entrainment of Volcanic Ash	2.44
2.3.8 Summary	2.44

2.4	EXPLOSIONS AND PRESSURIZED RELEASES	.	.	.	.	.	.	2.45
2.4.1	Dust Explosions	.	.	.	.	.	.	2.48
2.4.2	Industrial Process Releases	.	.	.	.	.	.	2.49
2.4.3	Nuclear Explosions	.	.	.	.	.	.	2.50
2.4.4	Fuel Cycle Explosions	.	.	.	.	.	.	2.53
2.4.5	Release Incidents	.	.	.	.	.	.	2.53
2.4.6	Coal Mine Explosions	.	.	.	.	.	.	2.56
2.4.7	High Explosive Releases	.	.	.	.	.	.	2.58
2.4.8	Natural Phenomena	.	.	.	.	.	.	2.58
2.4.9	Other Data	.	.	.	.	.	.	2.58
2.4.10	Experimental Investigations	.	.	.	.	.	.	2.59
2.4.11	Pressure Waves	.	.	.	.	.	.	2.61
2.4.12	Liquids	.	.	.	.	.	.	2.62
2.4.13	Post-Release Movement of Particles	.	.	.	.	.	.	2.66
2.4.14	Summary	.	.	.	.	.	.	2.66
3.0	COMMINUTION	.	.	.	.	.	.	3.1
3.1	INDUSTRIAL PROCESSES	.	.	.	.	.	.	3.2
3.2	SOLID WASTE INVESTIGATIONS	.	.	.	.	.	.	3.3
3.2.1	Breakup of Fuel Forms	.	.	.	.	.	.	3.11
3.2.2	Shock Compression	.	.	.	.	.	.	3.11
3.2.3	Comminution of Coal	.	.	.	.	.	.	3.12
3.2.4	Summary	.	.	.	.	.	.	3.14
4.0	BEHAVIOR OF LIQUID PARTICLES	.	.	.	.	.	.	4.1
4.1	LEAKS AND SPILLS	.	.	.	.	.	.	4.3
4.2	ENTRAINMENT OF DROPLETS	.	.	.	.	.	.	4.4

4.3	SPRAYS	.	.	.	.	.	.	.	.	.	4.5
4.3.1	Physical Processes	.	.	.	.	.	.	.	.	.	4.6
4.3.2	Size Distribution	.	.	.	.	.	.	.	.	.	4.7
4.3.3	Breakup	.	.	.	.	.	.	.	.	.	4.11
4.4	FLASHING SPRAYS	.	.	.	.	.	.	.	.	.	4.18
4.5	EVAPORATION	.	.	.	.	.	.	.	.	.	4.21
4.6	LIQUID POOL RELEASE	.	.	.	.	.	.	.	.	.	4.24
4.7	SPARGING	.	.	.	.	.	.	.	.	.	4.26
4.8	SUMMARY	.	.	.	.	.	.	.	.	.	4.27
5.0	CONDENSATION	.	.	.	.	.	.	.	.	.	5.1
5.1	ATMOSPHERIC CONDENSATION AEROSOLS	.	.	.	.	.	.	.	.	.	5.1
5.2	CONDENSING SYSTEMS	.	.	.	.	.	.	.	.	.	5.2
5.3	SPRAY CONDENSATION	.	.	.	.	.	.	.	.	.	5.3
5.4	STUDIES OF CORE DISRUPTIVE ACCIDENTS	.	.	.	.	.	.	.	.	.	5.3
5.4.1	Nuclei Formation--Category 1	.	.	.	.	.	.	.	.	.	5.3
5.4.2	Nuclei Formation--Category 2	.	.	.	.	.	.	.	.	.	5.4
5.5	OTHER CONDENSATION MODELS	.	.	.	.	.	.	.	.	.	5.4
5.6	GAS-TO-PARTICLE CONVERSION	.	.	.	.	.	.	.	.	.	5.5
5.7	SUMMARY	.	.	.	.	.	.	.	.	.	5.5
6.0	AIRBORNE CHEMICAL REACTIONS AND CONVERSIONS	.	.	.	.	.	.	.	.	.	6.1
6.1	HETEROGENEOUS PROCESSES	.	.	.	.	.	.	.	.	.	6.1
6.2	ADSORPTION	.	.	.	.	.	.	.	.	.	6.3
6.3	AEROSOL SURFACE AREA	.	.	.	.	.	.	.	.	.	6.3
6.4	SUMMARY	.	.	.	.	.	.	.	.	.	6.5
7.0	REFERENCES	.	.	.	.	.	.	.	.	.	7.1

FIGURES

2.1-1	Nominal PuO ₂ Particle Size Distribution	2.4
2.1-2	Mass Concentration of Solid Particles in Aerosols	2.9
2.3-1	Aerodynamic Entrainment of UO ₂ Powder from Various Surfaces	2.19
2.3-2	Diagram of Successive Stages of Dispersal of Cohesive-Type Dust Beds	2.29
2.3-3	Effects of Relative Humidity on Surface Particle Retention	2.39
2.4-1	Comparison of Cumulative Fractions of Organic Liquid Airborne for Log-Normal and Semilog Distributions	2.64
2.4-2	Energy Requirements for Subdivision and Dispersion of Liquids and Solids	2.65
3.2-1	Particle Size Distribution of Glass Crushed by Impact	3.7
3.2-2	Particle Size Distribution of Type III Concrete Crushed by Impact	3.7
3.2-3	Particle Size Distribution of High Alumina Concrete Crushed by Impact	3.8
3.2-4	Particle Size Distribution of Type I-P Concrete Crushed by Impact	3.8
3.2-5	Averaged Size Distribution Curves for Soda-Lime-Silica Glass Impacted at Four Different Energies	3.9
3.2-6	Size Distribution for Soda-Lime-Silica Glass Impacted by the Same Total Amount of Energy, 158.4 ft-lb, Applied as Single or Multiple Blows	3.9
4.1	Pressure Exerted on Drops by Various Mechanisms	4.18
4.2	Normalized Mean Drop Size as a Function of Spray Temperature	4.21
4.3	Calculated $h_{\max}$ in Evaporator	4.24
6.1	Comparison of Experimental Aerosol Surface Areas for a Given Rate of Aerosol Formation	6.4

TABLES

1.1	Classification of Aerosols	1.1
2.1-1	Dispersibility of Some Powders	2.3
2.1-2	Some Airborne Particulate Mass Concentrations	2.7
2.2-1	Resuspension Factors from Wind Resuspension Stresses	2.12
2.2-2	Resuspension Factors from Mechanical Resuspension Stresses	2.13
2.2-3	Resuspension Rates	2.15
2.3-1	Aerodynamic Entrainment of Uranium Particles in the Respirable Size Range from Various Surfaces	2.18
2.3-2	Suspension Fluxes from Various Surfaces, Mass Fraction/Sec	2.19
2.3-3	Fractional Release During Air Drying of Concentrated Plutonium Nitrate Solutions from a Plutonium Source	2.20
2.3-4	Calculated Entrainment Fluxes for Plutonium Nitrate from Stainless Steel, mass fraction/sec	2.21
2.3-5	Local Air Velocities for Dispersal of Dust Ridges	2.23
2.3-6	Local Air Velocities for Dispersal of Dust Beds	2.23
2.3-7	Entrainment Rates of <14 μ m Coal in Cohesive Dust Beds	2.24
2.3-8	Entrainment of Dust Beds Produced by Air Generated in an Explosion	2.37
2.4-1	Categories of Accidental Explosions and Where They Are Most Likely to Occur	2.46
2.4-2	Types of Explosions in a Chemical Industry Study, 1964 to 1968	2.47
2.4-3	TNT Pressure Equivalents	2.47
2.4-4	Relationship Between Dust Fines and Explosive Concentrations	2.48
2.4-5	Properties of Fallout Particles	2.51

2.4-6	Characteristics of Airborne Material from Project Schooner	2.52
2.4-7	Summary of Explosion Source Term Data from Environmental Reports	2.53
2.4-8	Mean Plutonium Size Characteristics of Aerosols at Five AEC Facilities Upstream from Air-Cleaning Facilities	2.54
2.4-9	Major Plutonium Explosions	2.55
2.4-10	Isotopic Composition of PuO ₂ Involved in Pressurized Container Rupture	2.56
2.4-11	Characteristics of Particles from Explosive Tests That Dispersed Depleted Uranium Metal	2.60
2.4-12	Comparison of Cumulative Fractions of the Mass of Airborne Material for Log-Normal and Semilog Size Distributions	2.63
3.1-1	Fineness of Materials Produced by Different Grinding Equipment	3.3
3.1-2	Particle Sizes Produced by Abrasive Cleaning	3.4
3.2-1	Particle Size Distribution of Glass Crushed by Impact	3.5
3.2-2	Particle Size Distribution of Concretes Crushed by Impact	3.6
3.2-3	Power Law Coefficients for Relating Airborne Respirable Dust Counts to Nonairborne Dust Counts	3.12
4.1	Forces on Droplets Expressed in Terms of Pressure	4.19
4.2	Fragmentation Drop Size Estimates	4.20
4.3	Evaporation Times for Water Droplets in Still Air at 20°C and P = 760 mm Hg	4.21
4.4	Life and Distance of Fall Before Complete Evaporation of Water Drops in Air for Wet Bulb Depression of 10°C	4.23
4.5	Life for Water Droplets in Air	4.23
5.1	Examples of Gas-to-Particle Conversion Mechanisms	5.6

5.2	Growth Laws for Gas-to-Particle Conversion	.	.	.	.	5.6
6.1	Values of k'_{expt} and ϕ for Selected Reactions	.	.	.	.	6.2

1.0 INTRODUCTION

Accidents in nuclear fuel cycle facilities could result in the airborne release of contained radioactive materials. Safety analysis reports and environmental impact statements require an estimation of these releases. At present it is difficult to write these assessments because information used in making the analysis is either nonexistent or scattered throughout the literature. The U.S. Nuclear Regulatory Commission's Division of Risk Analysis has sponsored this work so that a source term description for accident-generated aerosols will be more readily available.

This review discusses the characteristics of accident-generated particles to aid in quantifying their airborne release. Data on the dispersibility, quantities, fractional releases and behavior of powders and liquids are also summarized (when available).

Traditionally aerosols have been classified as condensation and dispersion aerosols (Fuchs 1964, pp. 1-2). Dispersion aerosols are formed by grinding of solids or by shear of liquids and resuspension of powder by air currents or vibration. Condensation aerosols are formed when supersaturated vapors condense or when gas-phase reactions result in a solid reaction product. These broad categories and subclasses have been tabulated (Dennis 1976, p. 12) and are shown in Table 1.1.

TABLE 1.1. Classification of Aerosols

<u>Condensation Aerosols</u>	<u>Dispersion Aerosols</u>	
<u>Liquid Particles (mists), Solid Particles (smokes), or Solid and Liquid Particles</u>	<u>Liquid Particles (mists)</u>	<u>Solid Particles (dusts)</u>
Examples:	Examples:	Examples:
Formation from superheated vapor, with or without foreign nuclei present	Nebulizer sprays	Grinding of solids
Gas phase--chemical reactions	Nozzle sprays	Pneumatic resuspension of powders
Photolysis products	Liquid shear	Blasting to shatter solids
Combustion products		Resuspension of solid particles from a liquid suspension

For the purpose of this work the traditional classification is reversed: the particle generation mechanism or source is identified and then the resulting aerosols are discussed, i.e., a generic stress such as grinding and its resultant aerosol are described. Each stress mechanism has a release factor to use in estimating releases.

Coagulation, agglomeration and similar phenomena are not within the scope of this literature review. Particle dynamics after an event are covered in a separate report which is in the scope of our study of accident aerosols.

Particle releases are caused by:

- Spills

For powder and liquid spills, limited information was found.

- Resuspension

There is a large body of information developed, and ongoing research is being conducted. Since this topic has been well-documented by other investigators, it is not covered in detail in this review. We have included release tables compiled by other reviewers, however.

- Aerodynamic Entrainment

Bureau of Mines studies have developed entrainment rate and pickup velocity equations. Some experimental work on aerodynamic entrainment, i.e., spills of coal into a moving airstream, have been included.

- Explosions and Pressurized Releases

Work has been done on large, nuclear-type events. No information was found on studies of limited releases of pressurized powders or liquids from containers.

- Comminution

Literature indicates that comminution of solids is a low-magnitude airborne release. No work on crushing or puncturing powder containers with subsequent release measurements was discovered. Breakup of hot or slurried material or cold surface impaction is another area with little available information.

- Liquid Particle Releases

These have been mentioned in conjunction with spills, sprays and explosions. A large body of information is continuing to be generated on liquid particles resulting from Loss-of-Coolant Accidents (LOCAs).

- Airborne Chemical Reactions

A large body of information exists on atmospheric reactions (particularly NO_x and SO_x). Some of this work showing the complexity of the phenomenon is contained in this review. Data on indoor pollution reactions were not searched for.

Sources reviewed were particle measurements made during or following accidents, bibliographies of aerosol behavior, models, and experimental measurements. The Department of Energy's (DOE's) Energy Information Data Base of bibliographic data was searched by computer. Because it is assumed that the reader is familiar with fundamental particle references, these are mentioned only briefly. Some effort was made to look at new information on related areas, such as the coal industry, where particle behavior is of concern.

2.0 BEHAVIOR OF PREFORMED PARTICLES

Mechanical stresses can generate airborne particles from preformed particle sources, which are powders or other materials in a divided form that can be physically dispersed in a release. This section discusses the releases from a source as the result of some physical action, such as powder falling from a ruptured container, producing aerosols, or particles resuspended from the spilled powder by air currents.

The magnitude of the releases from mechanical stresses must be evaluated. It is essential to identify the amount and particle size of the released particles, for they provide the source term for releases. The total airborne material generated establishes the severity of an event, while the aerosol particle size establishes the transport and inhalation potentials of the aerosol. Particle size provides the key to the relative hazard to the individual: those particles $<10\text{ }\mu\text{m}$ aerodynamic-equivalent diameter (AED) are easily inhaled and retained in the bronchial system. Because practically all inhaled particles $\geq 10\text{ }\mu\text{m}$ are deposited in the nasopharyngeal region, they have much less radiological significance than do smaller particles that are deposited preferentially in the tracheobronchial and pulmonary regions. Therefore, our discussion of releases of preformed particles includes methods of estimating the quantity and particle size of the releases where the information is available.

Under most circumstances, preformed particles, i.e., powders, are more easily injected into the air than other physical forms when subjected to equal force. Forces initiating releases are discussed in this section and include powder spills, resuspension, aerodynamic entrainment, and explosions and pressurized releases.

2.1 SPILLS

A spill of powder appears to be the simplest accident event. The spill could be caused by overturning an open vessel, resulting in a slug flow of the contained powder to the floor, or it could be in the form of a powder leak, resulting in a powder stream falling through air during a finite period of

time. Either event can involve varying unknown amounts of powder. The powder falls until it impacts on the floor surface of the enclosure, dispersing into the air as it falls, with some being entrained in the airflow. Airborne releases thus can occur as the powder is falling and be further scattered by the force of the impact. Spills of powder into a moving airstream are considered in Section 2.3, Aerodynamic Entrainment.

2.1.1 Discussion

Spills into static air are low release events. They could occur if operation of the facility ventilation system requires little airflow or it is interrupted as the result of an accident. Very little air movement would occur in storage facilities, for example.

While the rate of fall of individual particles can be calculated, Bagnold (1941) noted that even a collection of similarly-sized particles or grains will not have the same rate of fall. Since the particles involved in a spill will not fall at the same rate, it would not seem that traditional individual particle velocities would apply to releases from a falling mass. However, an estimate of the rate at which the material will fall can be made if the diameter of the leak is known. Schwendiman (1977) reported on gravity flow of sand through capillaries. The fine size fraction, 120 μm to 400 μm , was coarser than likely to be found in the mixed oxide (MOX) fuel plant materials. The gravity flow is a function of the diameter of the opening and can vary by as much as the 2 to 3.5 power of the diameter.

During a fall, powder disperses in the air. Fuchs (1964, pp. 372-373) has reviewed dispersal studies and found them extremely scarce. In the single set of experimental measurements located, he reported 2 cm^3 powder was poured through a narrow slit into a vertical tube 250 cm in height and 4.5 cm in diameter. The particles were separated to some extent as they fell through the air, and the percentage of powder which had not settled on the bottom of the tube in 6 sec was determined. In this time, the powder could not have attained its maximum dispersibility. The dispersibility which was determined is shown in Table 2.1-1.

TABLE 2.1-1. Dispersibility of Some Powders

Dust	Particle Radius Limits, μm	Dispersibility, Percent
Lycopodium	12	100
Wood charcoal dust	0 to 25	85
	0 to 7	23
Aluminum powder	0 to 15	66
Talc	0 to 20	57
Carbon black	0 to 15	47
Potato starch	0 to 35	27
Graphite dust	0 to 25	17
Pulverized slate	0 to 25	13
Cement	0 to 45	5.5
Prepared chalk	0 to 6	1.5
Polydisperse silica dust (coarse)	--	21
Polydisperse silica dust (fine)	--	8
	11.5	68
Isodisperse silica dust	8	83
	5.6	45
	7	50
Porcelain dust with fine frac- tions removed	2.7	52
	1.1	21
	0.45	12
Porcelain dust without fine fractions removed	--	5

These figures illustrate the anticipated range of powder dispersibility and the need to consider the kind of powder involved in a spill. In the nuclear fuel facility (e.g., an MOX plant), PuO_2 and MOX are the powders of concern. The size distribution could be similar to the PuO_2 size distribution reported by Schwendiman (1977) and shown in Figure 2.1-1. The mass fraction of material with a 10 μm AED is 0.1.

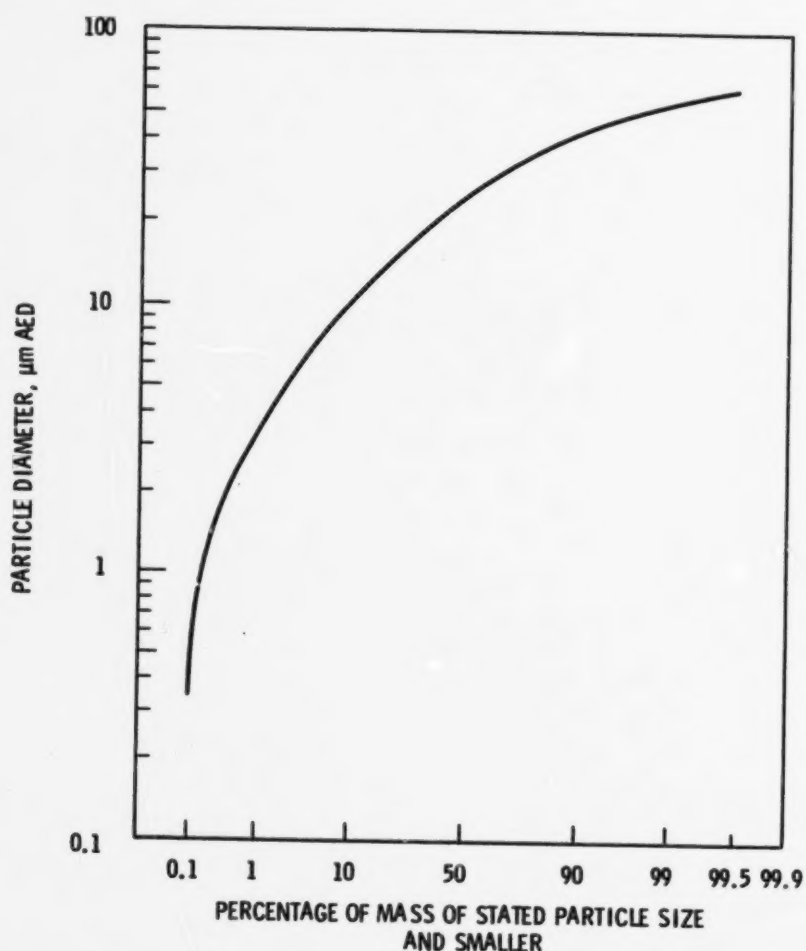


FIGURE 2.1-1. Nominal PuO₂ Particle Size Distribution

Unless the particles are quite small, as in tobacco smoke, true gas-phase dispersions of particles have only a transitory existence (Orr 1966, p. 106). Even when the particles are finely divided, as in aerosols, they are inherently unstable.

The powder's moisture content and temperature can potentially influence powder flow (e.g., powder spilling from an aperture). Moisture greater than the moisture in equilibrium with surroundings would generally enhance "stickiness" of the powder and prevent flow. However, some tests of powder flow in air made with mixtures of tungsten powder 8 to 16 μm dia showed optimum flow at 45 percent relative humidity (RH) (Hausner 1971, p. 25). Other powders would

require different relative humidities for optimum flow. In another study, using dry sand in gravity flow tests (Bingham and Wikoff 1931), temperature effects were not well defined but were considered minimal. Disaggregation from a powder spill, either the overturning of an open container or a continuous powder stream pouring from a breached container, could be influenced by the RH parameter.

Falling aggregates can be ruptured by air currents: a shearing force develops and increases with the size of the aggregate and with the relative velocity of the aggregate and the air (Fuchs 1964, p. 374). The larger the aggregate the more easily it ruptures when falling through air. The velocity of a fall is generally small, but increased disaggregation results when aggregates are introduced into a rapidly flowing airstream.

The downward force of gravity depends on the falling object and its density. The resisting force depends on the area of frontage exposed to the fluid, the shape of the object and speed through fluid (Bagnold 1941). A great deal of work has been devoted to the rate of fall of individual particles in a fluid at rest, and calculations have been developed (Dallavalle 1948). These calculations, however, are not considered appropriate for estimating spill releases because of frontage effects.

Impaction is another consideration in powder spills: the powder disperses as it falls and is further disaggregated on impaction with a surface when agglomerates are broken. If the powder particles are not monodispersed, the material could have larger host particles holding smaller particles on the surface, a situation similar to soil structure. If so, the submicron- and micron-sized particles will be shed from the surface of the larger host particle (Rosinski 1976). The numbers shed would be dependent on the size range of the parent particle: the greater it is, the more particles shed by surface impaction after free fall.

One study (Pring 1949) investigated falling material and air currents generated by the motion of the falling solids. The observations made on this phenomenon are of some interest:

- Compact solids are dropped, expand through the falling distance, then conglomerate upon impact at the bottom of a system. This was considered a bellows effect, with air that was filling voids between particles forced out by compacting material at the bottom of a drop.
- The force exerted by the falling particles creates a negative pressure within and about the material column as it drops. The differential between the zone of normal pressure outside the column and that of reduced pressure induces a flow of air toward and along with the falling solids.

In a laboratory study of the formation of airborne respirable dust created by dropping broken coal from a belt conveyor into a 77.6-cm/sec airflow, respirable dust adhering to the coal became airborne (Cheng 1973). The study used a polydisperse coal ranging up to 4 in. dia. Airborne dust increased with higher drops. The value for factor R, the mass airborne dust/unit mass of broken coal, was 10^{-3} in these studies. This comprised 10 percent of the respirable dust adhering to the larger pieces of coal.

Further observations on the behavior of the falling coal were made. When its fall is arrested, the mass of dust-laden broken coal suddenly compacts. Airborne dust is formed as the result of the impact shock, dislodging adhering dust from the surface. The surge of air originating from the space gaps in the impacting coal frees the dust from the impacting coal and secondary air currents carry it away. The void fraction was therefore determined to have a direct bearing on the specific formation of respirable dust.

The airborne concentration of particles within an enclosure may achieve high transitory levels followed by decay. The value of the airborne concentration could be used to estimate the magnitude of the release if the volume of the enclosure is known or if the concentration is maintained in an airstream of a known volume. Table 2.1-2 lists some airborne particulate mass concentrations that could be extrapolated to calculate airborne releases; some consideration must be given to the particle size distribution in the enclosure volume.

It has been observed that the particle size distribution of airborne aerosols in steady aerosol formation is constant or "self preserving" (ORNL

TABLE 2.1-2. Some Airborne Particulate Mass Concentrations

Aerosol	Mass Concentration	Reference
Dust storm	0.5 to 10 g/m ³	First 1952
Uranium dioxide powder	10 g/m ³ (5 sec) to 0.1 to 0.01 g/m ³	Schwendiman 1977
Dust devil	5 g/m ³	Sinclair 1974
Steam generating station	50 to 3000 mg/m ³	Bond 1972, p. 62
Mine working face (no control)	500 mg/m ³	First 1952
Mine air	0.05 to 0.5 g/m ³	First 1952
Foundry workroom	2 to 30 mg/m ³	First 1952
Los Angeles smog	0.5 to 50 mg/m ³	Bond 1972, p. 62
Nuisance dust	10 mg/m ³	United Power Assoc. 1974
Industrial atmosphere	0.1 to 50 mg/m ³	Dennis 1976, p. 9
Cigarette smoke (steady state, cocktail party)	5 mg/m ³	Stern 1976, p. 157
Ambient atmosphere	0.05 to 1.0 mg/m ³	Dennis 1976, p. 9
Air conditioned building	0.3 mg/m ³	First 1952
Cigarette smoke (average)	40 to 400 µg/m ³	Stern 1976, p. 157

1970). Small particles tend to agglomerate rapidly by Brownian motion, while large particles are removed by impingement or sedimentation. It has been suggested that a quasi-stationary state exists so that the rate at which matter enters a differential size is equal to the rate at which matter is lost by sedimentation. It has been determined that the stable concentration of particles less than ~3 µm dia is consistently less than a few g/m³ after the aerosol has been permitted to age a few seconds or minutes. A formulation for

the differential concentration of particles in a metastable aerosol as a function of size relative to the initial concentration, size, and a half-life of particles is:

$$C_0 = \frac{\pi D_0^3 \rho}{6 K t_{1/2}}$$

where

C_0 = initial concentration

D_0 = diameter of the agglomerate, μm

ρ = density of the agglomerate, g/cm^3

K = agglomeration coefficient,

$\approx 9 \times 10^{-11} \text{ cm}^3/\text{sec}$ for $D_0 \leq 0.3 \mu\text{m}$

$\approx 3 \times 10^{-10} \text{ cm}^3/\text{sec}$ for $D_0 \geq 0.3 \mu\text{m}$

$t_{1/2}$ = time since beginning of agglomeration, half-life

It has been found that agglomerates (even those of dense particles) have a density of 1 g/cm^3 . Assuming a half-life of 10 min and converting to appropriate units, this expression becomes:

$$C_0 = 9700 \rho D_0^4 \quad \text{where } D_0 \leq 0.3 \mu\text{m}$$

and

$$C_0 = 2900 \rho D_0^3 \quad \text{where } D_0 \geq 0.3 \mu\text{m}$$

These expressions for aerosol concentration are shown for a wide variety of heavily concentrated and turbulent aerosols (smoke, fly ash, and dioctylphthalate in air) in Figure 2.1-2. For concentrations of monodispersed aerosols with a half-life of 10 min, these expressions provide a practical upper limit for the concentration of solid particles in air (ORNL 1970). Values from this plot could be used for calculating airborne releases/ m^3 of air after evaluating the postulated accident conditions.

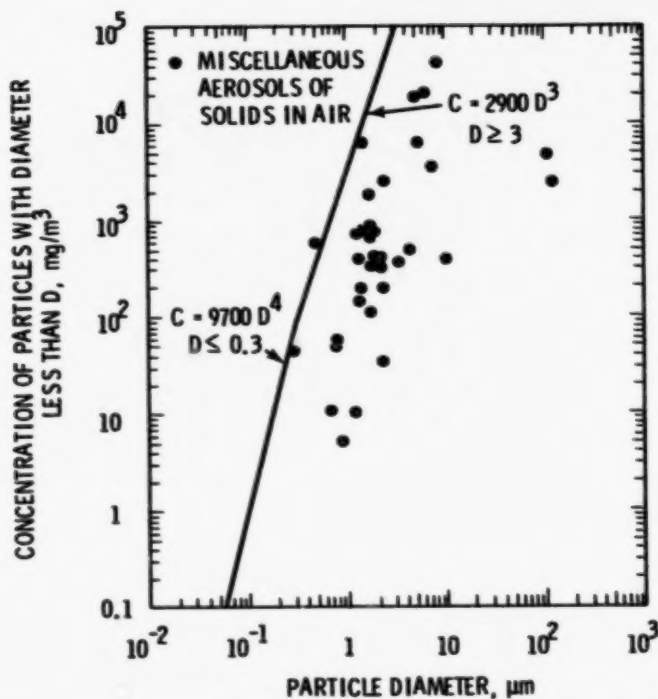


FIGURE 2.1-2. Mass Concentration of Solid Particles in Aerosols

2.1.2 Summary

None of the literature cited really confronts the following problems associated with estimating accidental releases from a free-fall spill:

- Particle spill can be influenced by frontage effects on bulk mass and subsequent impact.
- The airborne particle is a transitory phenomenon.
- A spill event does not produce a homogeneous aerosol.
- More airborne material would be anticipated in the immediate vicinity of a spill.

2.2 RESUSPENSION

Airflow over surface deposited materials can reentrain particles, making them airborne in a process known as resuspension. Resuspension in a facility issues from lightly fixed surface contamination deposited on exposed surfaces

by natural mechanisms during the facility's operation (the contamination is then removed incidentally by forces generated in an accident), or they can be freshly deposited during an accident and subsequently removed by resuspension forces. The amounts that become airborne can be estimated using resuspension factors, resuspension rates, or fractional releases. Mathematical models proposed to estimate aerodynamic stresses, e.g., air stresses on piles of powder, will be discussed in Section 2.3.

2.2.1 Resuspension Factors

Resuspension factors are defined as the airborne concentration/ m^3 of air divided by the surface contamination/ m^2 below the airborne measurement (Sehmel 1979). This relationship is called K and is described by the expression

$$K/m = \frac{\text{airborne contamination}/m^3}{\text{surface contamination}/m^2}$$

Sehmel considered resuspension factors most useful for predicting concentrations of airborne material above contaminated surfaces; however, he noted they are not useful in predicting changes with time in surface contamination levels. Thus the resuspension factor can be used to calculate the airborne contamination. To be useful for estimating the total release, estimates of the affected volume must be made (i.e., if in an enclosure, estimates of how much of that enclosure contains airborne contamination). If the release is in a known airflow, it is assumed uniformly distributed in the airflow.

To make a source term estimate, some knowledge of the surface contamination is required:

- quantity
- type
- firmness of its fix
- amount of resuspension
- source continuity or finiteness
- anticipated duration.

A threshold value of the air-caused surface stress on a particle must be exceeded before a particle is resuspended (Bagnold 1941). This threshold stress is a function of particle size as well as surface properties. Bagnold's soil movement studies showed that 90 percent of the airborne soil mass moves within 1 ft of the surface and contains relatively large particles. Particles above $\sim 100 \mu\text{m}$ dia would be carried aloft only if wind speeds become significantly larger than the lowest wind speed required to produce soil movement. For particle size mixtures, the threshold speed is smaller than that required to erode only the largest particle.

The maximum dust concentration measured in a dust devil was $\sim 5 \text{ g/m}^3$ (Sinclair 1976). This is a value that might be used to estimate the maximum airborne powder subsequent to an event with severe wind stresses.

In an accident, it seems that wind resuspension and/or mechanical stress resuspension could be involved. Resuspension factors for wind resuspension and from mechanical stresses applicable to outdoor and indoor conditions have been compiled by Sehmel (1979) and Walker (1978) and are shown in Tables 2.2-1 and 2.2-2. It can be seen that the resuspension factors can cover a wide range of values from $10^{-2}/\text{m}$ for vigorous sweeping to $10^{-11}/\text{m}$ for a liquid solution on cleared sandy soil. In evaluating the accident, the resuspension factors and stresses are considered, leading to a selection of the most appropriate.

A resuspension factor can then be calculated:

$$\text{Surface contamination}/\text{m}^2 \times \text{resuspension factor}/\text{m} = \text{airborne material}/\text{m}^3.$$

It can be seen that resuspension factors must be used with caution and based on a careful assessment of the source material and the event making the material airborne.

2.2.2 Resuspension Rates

Resuspension rates are defined as the fraction of a source material airborne over a period of time.

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TABLE 2.2-1. Resuspension Factors from Wind Resuspension Stresses

Location	Source Material	Wind Speed Dependency	Resuspension Factor Range, m^{-1}	Reference
Nevada test site, Schooner	^{131}I		10^{-7} to 6×10^{-5}	Anspaugh et al. 1970
Nevada Test Site, GMX near center	Pu		3×10^{-10}	Anspaugh, Shinn and Phelps 1975
GMX near edge			3×10^{-9}	
New York—average air concentrations and surface depositions	Fallout		5×10^{-9} to 2×10^{-6}	Calculated from Bennett 1976
New York	^{238}U		5×10^{-8}	Bennett 1976
United Kingdom	^{238}U		5×10^{-9}	Bennett 1976
Palmares, Spain	Pu 10 day samples	$u < 9.7$ m/sec	1.4×10^{-9} to 7.8×10^{-6}	Calculated from Icanzo and Salvador 1970
Nevada test site, dusty rural air	Pu		7×10^{-6}	Langham 1971
Hanford, Washington	Inert tracer, 1 m	Increases	1.7×10^{-10} to 2.7×10^{-7}	Sehmel and Lloyd 1975
Rocky Flats, Kentucky	Pu		10^{-9} to 10^{-5}	Sehmel and Orgill 1973
Maralinga trials	U 0.3 m ht 0.6 m ht	$u < 5$ m/sec	3×10^{-4}	Stewart 1967
Paving stones	U_{308} , 2 m^2 source	$u < 5$ m sec	8×10^{-9} to 6×10^{-6}	Stewart 1967
Sandy desert grass	U_{308} , 9 m^2 source		1×10^{-9} to 5×10^{-5}	
Sandy soil with charred debris	^{210}Po , 1 m^2 source	$u < 5$ m/sec	2×10^{-9} to 9×10^{-6}	Stewart 1967
	^{210}Po , 1300 m^2 source	$u < 5$ m/sec	7×10^{-8} to 1×10^{-5}	
Monte Bello Islands, hurricane trials	Fallout: 16 m source, lightly vegetated sand and rock near tower. Shot near road, no disturbance	$u < 5$ m/sec	1×10^{-6} to 1×10^{-5} 1×10^{-8} to 2×10^{-7} 1×10^{-8} to 1.5×10^{-6}	Stewart 1967
Cleared sandy soil	YCl_3 , aqueous, 1 m^2 source	$u < 5$ m/sec	9×10^{-11} to 5×10^{-8}	Stewart 1967
	YCl_3 , aqueous, 9 m^2 source		5×10^{-10} to 6.2×10^{-7}	

TABLE 2.2-2. Resuspension Factors from Mechanical Resuspension Stresses

Location	Source Material	Resuspension Stress	Resuspension Factor Range, m^{-1}	Reference
Unventilated room	Be	Vigorous sweeping	1×10^{-2} to 4×10^{-2}	Mitchell and Sutsler 1967
Small unventilated room	Alpha floor contamination	People walking	3×10^{-4} to 2×10^{-2}	Calculated from Brunskill 1967
Change room with no ventilation	Alpha on coveralls	2 to 4 people changing coveralls	2×10^{-4} to 1.7×10^{-2}	Calculated from Brunskill 1967
Change room, concrete floor, 9 air changes/hr	"Loose" alpha floor contamination	4 to 6 people moving in room	2×10^{-4} to 3×10^{-3}	Brunskill 1967
Simulated work	Asbestos contaminated coat	Machining operations	1.2×10^{-3} to 5.3×10^{-3}	Carter 1970
		Stacking sheets	2.0×10^{-3} to 4.2×10^{-3}	
Room-concrete floor	Pu facility	No circulation	1×10^{-3} to 2×10^{-4}	Glauberger, Bootmann and Breslin 1967
		Fan air stress	3×10^{-4} to 3×10^{-3}	
		Fan and dolly movement	4×10^{-3} to 1.5×10^{-2}	
		No circulation after tests	5×10^{-4} to 1×10^{-3}	
Room-concrete floor	U facility	No circulation	7×10^{-5} to 4×10^{-4}	
		Fan air stress	3×10^{-5} to 2×10^{-4}	
		Dolly movement	1×10^{-4} to 2×10^{-4}	
		Fan and dolly movement	2×10^{-4} to 1×10^{-3}	
Maralinga trials	Fallout	Road survey, at 1-2 days	1×10^{-8} to 2×10^{-6}	Stewart 1967
		Cab Land Rover, 5th hr	6.4×10^{-5}	
		8th hr	2.5×10^{-5}	
	U	Dust stirred, 0.3 m ht	1×10^{-3}	Stewart 1967
	Pu	Vehicle dust, 0.3 m ht	3×10^{-7} to 7×10^{-4}	
		Pedestrian dust, 0.3 m ht	1.5×10^{-6} to 3×10^{-4}	
Particles settled in room	ZnS	Vigorous work-sweeping	1.9×10^{-4}	Fish et al. 1967
	MMD = $3.1 \mu m$	Walking	3.9×10^{-5}	
	$\sigma_g = 1.9$	Light work	9.4×10^{-6}	
	CuO	Light sweeping	7.1×10^{-4}	Walker 1978
	MMD = $2.0 \mu m$, $\sigma_g = 2.3$, alpha particulate	Pedestrian and equipment	4.6×10^{-3} to 5×10^{-5}	
		Bomb recovery operation with brick/plaster dust		
Civil defense trial	131I	Enclosed space	4×10^{-5} to 2×10^{-4}	Stewart 1976
		Open area	2×10^{-6}	

TABLE 2.2-2. (continued)

Location	Source Material	Resuspension Stress	Resuspension Factor Range, m^{-1}	Reference
Nevada test site	Pu	Extensive vehicular traffic	7×10^{-5}	Langham 1971
Laboratory room	PuO ₂	No movement	2×10^{-8}	Jones and Pond 1967
		14 steps/min	10^{-6}	
		36 steps/min	5×10^{-6}	
Monte Bello Islands, hurricane trails	Fallout	Road survey from back		Stewart 1967
		Land Rover, 4th day	8×10^{-7} to 3×10^{-5}	
		7th day	7×10^{-7} to 4×10^{-6}	
Australian desert "Totem" 1953	Fallout gamma	At tailboard, 7th day	1.6×10^{-5} to 3.1×10^{-5}	Stewart 1967
		Walking survey	3×10^{-7}	
		Vehicle survey at tailboard	2×10^{-6}	
Contaminated field	Pu	Inside vehicle	2×10^{-5}	Calculated from Milham et al. 1976
		Downwind tractor	5×10^{-8} to 1.0×10^{-6}	
		in tractor cab	2.4×10^{-7}	
		Fertilizing	7.9×10^{-9}	
		Subsoiling	5×10^{-6}	
		Planting, light discing	1.8×10^{-8} to 7.9×10^{-6}	
		Subsoiling	5×10^{-6}	
		Planting, light discing	1.8×10^{-8} to 7.9×10^{-6}	
		Mowing	1.8×10^{-5}	
Tractor cab in contaminated field	Pu	Tractor operations	4×10^{-9} to 2×10^{-6}	
New York	Fallout Pu	Average air concentration/surface concentration	5×10^{-9} to 2×10^{-6}	Calculated from Bennett 1976
Palmares, Spain	Pu	Yearly average farming	1.2×10^{-10} to 8.8×10^{-7}	Calculated from Iranzo and Salvador 1970
Contaminated sludge	Pu	Rototilling	5.6×10^{-8}	Calculated from Myers et al. 1976

Resuspension rates for outdoor situations were calculated or measured. Transport and diffusion models interpreting measured concentrations at only one height led to calculated rates. Others were measured and developed by integration of airborne concentrations as a function of height (Sehmel 1979). Values are shown in Table 2.2-3. The lower half of the table shows measured resuspension rates. To use them requires an estimation of the source area involved in the resuspension.

2.2.3 Fractional Releases

Under accident conditions a portion of the material can immediately become airborne. A fractional release can be calculated, i.e., airborne mass/source mass, if the information is available. Experimental measurements of fractional releases have been made for more vigorous aerodynamic entrainment stresses (Mishima 1975) and are discussed in Section 2.3.

TABLE 2.2-3. Resuspension Rates

Location	Source Material	Resuspension, sec^{-1} , Determined from:		Reference
		Calculation Using Transport Mode and Concentration at Only One Height	Integration of Measured Concentrations	
Corn leaves	Pollen		10^{-6} to 10^{-4}	Calculated from Aylor 1976
Nevada test site	Pu	2.7×10^{-12} to 4.8×10^{-10}		Anspaugh, Shinn and Phelps 1975
Nevada test site, GMX	Pu	10^{-12} to 10^{-10}		Healy 1974
Nevada test site	Pu	1×10^{-10} to 6×10^{-8}		Healy 1974, calculated from Wilson, Thomas and Stannard 1960
Hanford site	ZnS	1×10^{-8} to 3.5×10^{-6}		Healy 1974, recalculated from Healy 1955 and Healy and Fuquay 1958
Eroding field	Soil			Calculated from Skidmore and Woodruff 1968
		2×10^{-9}		
		2×10^{-7}		
		1×10^{-6}		
Prairie terrain	Calcium molybdate	10^{-8} to 10^{-10}		Sehmel and Lloyd 1975
Hanford site	Respirable particulates	10^{-11} to 10^{-7}		Sehmel 1979

2.2.4 Proposed Models

Wind stresses on individual sand grains have been modeled. Force balances including frictional drag, form drag, gravitation, and Bernoulli effect were considered (Akiba 1975; Andres 1972; Chepil 1950, 1959, 1965; Ford 1957; Kawamura 1975; Malina 1941; and Slinn 1976).

Agglomerate rather than individual particle resuspension would be more likely in estimating accident releases. Such resuspended toxic aerosol was modelled by Porph (1979) using a mass-consistent wind field model (MATHEW) combined with a particle-in-cell diffusion and transport code (ADPIC). He suggested an hourly averaged maximum resuspension factor of $10^{-4}/m$ for a 15 m/sec wind. He developed computer codes which interface with MATHEW-ADPIC numerical models to estimate the effect of resuspension of deposited toxic material.

2.2.5 Particle Size

Dusts which are produced by the redispersion of settled particles are usually highly aggregated (Green 1957). Gillette (1976) found that the breakup of host soil aggregates by wind was a function of soil texture. Production of particles with radius r of $<10 \mu m$ by soil wind erosion increased with wind speed at about the same rate as the horizontal sand flux for coarse soil. However, this rate was increasingly greater for fine-textured soils as the wind speed increased. Gillette attributed this to breakup of soil aggregates by sand blasting, but he did not present calculations to describe this breakup.

Powders that might be involved in fuel cycle accidents are often so fine that they might exhibit enhanced small particle production.

2.2.6 Summary

As can be seen from this review, considerable work has been done on resuspension and it is the subject of ongoing research. Resuspension rates range from 10^{-12} to 10^{-4} fraction/sec and resuspension factors from 10^{-10} to $10^{-2}/m$. In an accident, material can be resuspended by a variety of stresses. To use either resuspension rates or factors to estimate releases, resuspension from stresses similar to the situation under consideration should be used.

2.3 AERODYNAMIC ENTRAINMENT

Aerodynamic entrainment of particles may be considered an energetic form of suspension, which is defined as the transport of material from a surface to the atmosphere (Sehmel 1975). This material may be particles on the ground surface which have lost their identity by becoming attached to host particles. In contrast to the less severe resuspension stresses, particles become airborne by aerodynamic entrainment as the result of severe stresses. An example is powder falling through an airstream--the particles immediately interface with the moving air, a vigorous stress. A high percentage of the material, perhaps 10 percent or more, can become airborne, as opposed to a less severe resuspension rate. The amount of the source airborne will depend on the intensity of the stresses and the nature of the particles. In general, wind is the resuspension force and the fractional removal rate can be fairly low, 10^{-8} /sec to 10^{-10} /sec, depending on the surface and wind speed.

The following experimental investigations have considered aerodynamic entrainment.

2.3.1 Aerodynamic Entrainment of Surface Contamination

Mishima and Schwendiman (1973) have reported the suspension of UO_2 powder and uranylhexahydrate (UNH) solutions from various surfaces at two air velocities in a wind tunnel. The data are tabulated in Table 2.3-1. Under the conditions of these experiments, the suspension of material is not linear with time, as shown in Figure 2.3-1. For UO_2 powder from sandy soil, a large fraction was suspended within 1 hr at 20 mph and within 8 hr at 2.5 mph, with little or no suspension for the remainder of the 24-hr sampling period.

Suspension fluxes from various surfaces, i.e., the mass fraction of UO_2 $<10 \mu m$ AED/sec, were calculated and are shown in Table 2.3-2. Most of the values shown are greater than the 10^{-8} /sec long-term resuspension rate.

Mishima, Schwendiman and Radasch (1968) measured the plutonium entrained in air drawn across concentrated plutonium solutions (250 g Pu/l) held in a stainless steel dish at velocities up to 100 cm/sec and at temperatures up to $100^\circ C$ (see Table 2.3-3). The data, recalculated as entrainment fluxes assuming a linear rate, are shown in Table 2.3-4.

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TABLE 2.3-1. Aerodynamic Entrainment of Uranium Particles in the Respirable Size Range from Various Surfaces

Surface	Uranium Dioxide Powder				Uranium Nitrate Solution			
	2.5 mph		20-23 mph		2.5 mph		20-23 mph	
	Percent Airborne	Hr Collected	Percent Airborne	Hr Collected	Percent Airborne	Hr Collected	Percent Airborne	Hr Collected
Smooth, sandy soil	0.24	6	1.7	24	0.0051 ^(a)	24	0.20 ^(a)	24
	0.023	24	9.8 ^(b)	24	0.0042 ^(a)	24	0.70 ^(a)	24
	0.005	24 ^(b)	0.68 ^(b)	24	0.037	6	0.027	28
					0.010	24		
Vegetation cover	0.0038	24	0.4	24	—	—	—	—
Stainless steel	0.075	4.8 ^(b)	1.1	24	0.017 ^(b)	6	0.78	5
			0.29 ^(b)	24				
Asphalt	0.087 ^(b)	6	—	—	0.033 ^(b)	6		

(a) Solid residues from air-dried UNH solutions.

(b) Solid residues remaining after a gasoline fire.

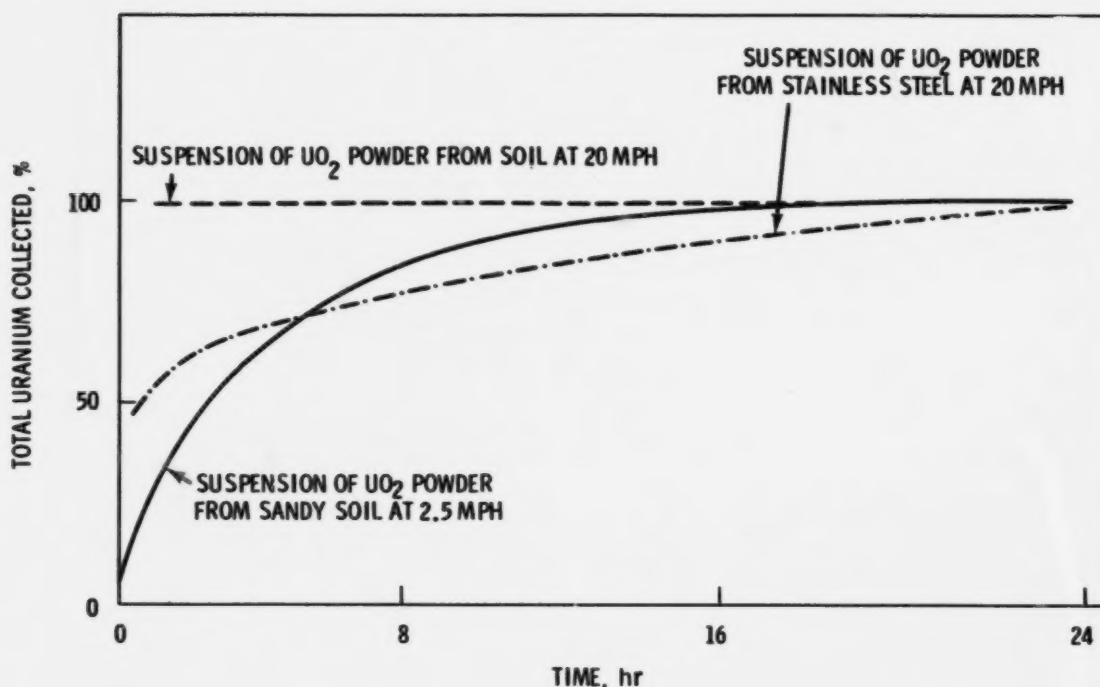


FIGURE 2.3-1. Aerodynamic Entrainment of UO₂ Powder from Various Surfaces

TABLE 2.3-2. Suspension Fluxes from Various Surfaces, Mass Fraction/Sec

Surface	Uranium Dioxide Powder		UNH Solution	
	2.5 mph	20 mph	2.5 mph	20 mph
Smooth, sandy soil	6.7×10^{-6}	1.2×10^{-5}	1.1×10^{-6}	1.6×10^{-7}
	1.6×10^{-7}	6.8×10^{-5}	6.9×10^{-8}	$1.4 \times 10^{-6(a)}$
	2.5×10^{-8}	4.7×10^{-6}	$3.5 \times 10^{-8(a)}$	$4.9 \times 10^{-6(a)}$
			$2.9 \times 10^{-8(a)}$	
Vegetation cover	2.6×10^{-8}	2.8×10^{-6}	--	--
Stainless steel surface	2.6×10^{-6}	7.6×10^{-6}	--	2.6×10^{-6}

(a) Solid residues from air-dried UNH solutions.

TABLE 2.3-3. Fractional Release During Air Drying of Concentrated Plutonium Nitrate Solutions From a Plutonium Source

Run Number	Temperature, °C	Run Velocity, cm/sec	Sampling Time, hr		Weight % Plutonium Found In:			
			Evap.	Residue	Containment Vessel Wash	Condensate Plus Wash	Sweep Air During Evaporation	Sweep Air Following Evaporation
N1(a)	Ambient	10	24	—	3.3×10^{-3}	8.7×10^{-3}	$<10^{-7}$	—
N2(a)	75	10	5	20	2.7×10^{-4}	9.5×10^{-8}	$<10^{-6}$	$<10^{-6}$
N3(b)	100	10	2	4	4.6×10^{-3}	1.7×10^{-6}	10^{-3}	3×10^{-7}
N4(b)	Ambient	50	24	24	3.5×10^{-4}	4.5×10^{-7}	2.5×10^{-7}	1×10^{-7}
N5(b)	100	50	1.5	3	2.7×10^{-2}	1.4×10^{-4}	3×10^{-3}	6×10^{-7}
N6(b)	90	50	2	4.5	5.1×10^{-4}	5.4×10^{-6}	5.3×10^{-5}	1×10^{-7}
N7(b)	Ambient	100	24	24	2.0×10^{-2}	7.5×10^{-8}	$<2 \times 10^{-8}$	$<2 \times 10^{-8}$
N8(b)	50	100	2	4	4.5×10^{-4}	9.4×10^{-6}	1.3×10^{-5}	$<2 \times 10^{-8}$
N9(b)	90	100	1.5	4	1.3×10^{-4}	9.4×10^{-5}	5.7×10^{-5}	3×10^{-6}

(a) 0.86 g plutonium used during these runs.

(b) 0.72 g plutonium used during these runs.

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TABLE 2.3-4. Calculated Entrainment Fluxes for Plutonium Nitrate from Stainless Steel, mass fraction/sec

Air Velocity, cm/sec	Temperature, °C	Plutonium Nitrate Solution	Air-Dried Residue from Plutonium Nitrate Solution
10	Ambient	$<6.9 \times 10^{-13}$	$<8.3 \times 10^{-12}$
	75	$<3.3 \times 10^{-11}$	$<1.2 \times 10^{-11}$
	100	8.3×10^{-8}	—
50	Ambient	1.7×10^{-12}	6.9×10^{-13}
	90	4.4×10^{-4}	3.7×10^{-11}
	100	3.3×10^{-7}	3.3×10^{-11}
100	Ambient	$<1.4 \times 10^{-13}$	$<1.4 \times 10^{-13}$
	50	1.0×10^{-9}	$<8.3 \times 10^{-13}$
	90	3.8×10^{-7}	1.2×10^{-10}

Selecting an appropriate suspension flux for accident assessment is difficult. The material could be distributed on a variety of surfaces (metal, concrete, soil with/without vegetation, etc.). The roughness of the surfaces can vary greatly (smooth concrete slabs to very coarse rubble), providing varying degrees of shielding for the deposited material. If the deposited material is buried under debris and equipment, the quantity suspended could be negligible.

2.3.2 Aerodynamic Entrainment of Coal Dust

The Bureau of Mines has sponsored experimental studies leading to calculations for estimating the entrainment of deposited coal dust (Singer, Cook and Grumer 1972; Singer, Harris and Grumer 1976; Hwang 1974). Tests were performed using cohesive dusts with particle diameters 6 to 80 μm , and free-flowing dust 100 to 150 μm . The work reviewed covered a wind stress range from minimum air velocity and dispersion rates to explosion-induced air flow. Some of the calculations developed may be limited to the situation under investigation. They are included mainly to enable understanding of the factors involved in aerodynamic entrainment.

2.3.2.1 Minimum Air Velocities and Entrainment Rates

These investigations (Singer, Cook and Grumer 1972) were designed to model some of the conditions that may develop in mines following a relatively weak or marginal gas explosion. They showed that 1) fresher deposits are more readily dispersed, 2) deposits are cohesive when compacted slightly, and 3) dusts larger than 100 μm disperse by different mechanisms than cohesive dusts.

It was found that a universal dispersion model did not appear feasible because of the large number of interdependent parameters in a mine at the time of an explosion. They were able to establish the effect of some of these parameters and to develop semiempirical equations relating minimum air velocities and entrainment rates with coal dust-bed properties such as particle size, compaction and location. Dust configurations used were ridges, flat-topped beds, semicylinders or cones. These experiments were performed in a wind tunnel 300 cm long, 7.62 cm wide, and 2.54 or 5.08 cm high.

For our study, the $<14 \mu\text{m}$ particle dia is considered the most appropriate figure to look at, since this size is similar to that of the cohesive powders found in nuclear fuel cycle facilities. However, these figures do not necessarily apply to nuclear fuel cycle powders. Tables 2.3-5 and 2.3-6 show the local air velocities for dispersal of dust ridges and beds, respectively. The slightly compacted landfill beds required a slightly higher air velocity for dispersal than ridges did. The apex angle showed the same trend with variations probably due to bed height.

Regression analysis related minimum air velocity for dispersion to dust-bed geometry (ridges or beds), cohesion, porosity, and bulk density porosity. Entrainment rates for the $<14 \mu\text{m}$ coal dust are listed in Table 2.3-7. Also, regression analysis of the data from cohesive dust beds^(a) of coal and rock dust was used to develop an equation (relative error 30 percent) for entrainment of cohesive dust beds:

$$E_c = 1.2 U_t^{0.33} \rho_B^{0.61} v^{0.76}$$

(a) Beds were 5.1 cm long x 2.5 or 5.1 cm wide x 0.32 or 0.54 cm high with perpendicular sides.

TABLE 2.3-5. Local Air Velocities for Dispersal of Dust Ridges

Material	Bulk Density, g/cm ³	Type of Compaction	Local Air Velocities, m/sec at Ridge Height and Apex Angle				
			0.64 cm, 120°	0.32 cm, 120°	0.16 cm, 120°	0.64 cm, 90°	0.64 cm, 60°
<14 μ m Pittsburgh seam coal Do(a)	0.43	Hand-Fill	10.3	9.8	8.4	7.1	4.7
	0.31	Free-Fall	7.1	5.5	4.0	5.6	4.3

(a) Explanation not available.

TABLE 2.3-6. Local Air Velocities for Dispersal of Dust Beds

Material	Bulk Density, g/cm ³	Bed Length, cm	Local Air Velocities, m/sec at Bed Height and Leading Angle at Base					
			0.32 cm, 90°	0.32 cm, 60°	0.32 cm, 30°	0.64 cm, 90°	0.64 cm, 60°	0.64 cm, 30°
<14 μ m Pittsburgh seam coal	0.43	2.5	5.9	8.2	10.3	8.9	12.1	12.6
		5.1	7.6	8.6	10.1	11.6	13.6	16.2

TABLE 2.3-7. Entrainment Rates of <14 μm Coal in Cohesive Dust Beds (perpendicular sides)

Size of Bed (LxWxH), cm	Velocity at Midheight, m/sec	Weight Removed, g	Time, sec		Entrainment Rate, g/sec	
			Total	Active	Total	Active
5.1 x 5.1 x 0.64	13.1	6.63	1.25	0.35	5.3	19
5.1 x 5.1 x 0.64	11.9	6.40	1.30	0.35	4.9	18
5.1 x 5.1 x 0.32	8.8	3.61	1.3	0.3	2.8	12
5.1 x 5.1 x 0.32	8.1	2.94	1.0	0.4	2.9	7.4
5.1 x 5.1 x 0.32	12.6	3.59	1.05	0.25	3.4	14.4
5.1 x 2.5 x 0.64	8.8	3.42	1.05	0.30	3.3	11.4
5.1 x 2.5 x 0.64	18.5	3.23	0.65	0.25	20	19

where

E_c = entrainment rate of cohesive dust bed, g/sec

U_L = velocity at midheight of dust bed, m/sec

ρ_B = bulk density, g/cm³

V = volume of dust bed, cm³.

Equation coefficients differed for dispersal of ridge-topped^(a) and flat-topped beds. There are three empirically developed equations for minimum air velocities to entrain flat-topped cohesive coal and rock dust beds (height <0.64 cm and length <5.1 cm) based on different input parameters relating to cohesion, porosity, and bulk density:

Ridges

$$U_L = 43.6 L^{0.26} A^{-0.39} \rho_B^{0.66}$$

$$U_L = 4.9 L^{0.26} A^{-0.42} \rho_B S_c^{0.29}$$

$$U_L = 32.4 L^{0.26} A^{-0.39} \left(\frac{1-\epsilon}{\epsilon} \right)^{0.12}$$

(a) With crest or leading angle normal to air flow.

Beds

$$U_L = 118 L^{0.26} H^{0.52} A^{-0.26} \rho_B^{0.67}$$

$$U_L = 3.61 L^{0.24} H^{0.59} A^{-0.26} \rho_B S_C^{0.47}$$

$$U_L = 96.6 L^{0.26} H^{0.52} A^{-0.27} \left(\frac{1-\epsilon}{\epsilon} \right)^{0.33}$$

where

U_L = air velocity at mid-height of dust bed, m/sec

L = bed length, cm

H = bed height, cm

A = angle at leading edge, deg.

ρ_S = solid density, g/cm³

ρ_B = bulk density, g/cm³

$\epsilon = \left(1 - \frac{\rho_B}{\rho_S} \right)$ = bed porosity

S_C = yield shear stress of bed material, dyne/cm².

Local minimum velocities for all cohesive deposits, independent of wind tunnel size, ranged from 5 to 30 m/sec. These values were considered suitable for use as a rough approximation of the range of minimum air velocities capable of dispersing coal and rock dust in any passageway. A Froude-type number, inertial force/restraining force FR_S (dimensionless), was used to relate minimum air velocities and dust bed properties. The fluid wall shear stress, $\tau_S = \rho_F (U_\star)^2$, is the same as the inertial force. The restraining force is due to a combination of gravitational and cohesive properties of the bed.

The entrainment behavior of cohesive deposits (rupture and removal of large clumps, then dispersal in midstream) appeared to depend on quite erratic events. Entrainment rates were not strongly dependent on bulk density or flow velocity, but on bed dimensions including height and bed length parallel to the direction of flow.

Photography was used to illustrate the mechanisms involved in entrainment of cohesive dust ridges and showed this sequence of events:

1. single-particle detachment of loose surface dust
2. removal of small clumps and particles
3. partial fracturing and subsequent entrainment
4. ridge breakup and complete entrainment of large clumps
5. continued breakup and dispersal of clumps in midstream.

Steps 1 to 4 characteristically required 100 to 500 msec to entrain 2-gram cohesive dust ridges.

2.3.2.2 Turbulent Gas Entrainment of Dust

The Bureau of Mines (Hwang 1974) formulated a theoretical model for calculating concentration of entrained dust from a dust deposit as a function of time. Calculations assumed steady air flow entrainment of eroded dust in the shock tube, considering it to be a turbulent diffusion process. The dust layer on the floor acts as a source, releasing a stream of dust, which mixes with the airstream. The intensity of the source, as well as diffusion coefficients, must be determined by fitting the theoretical solution with experimental results.

Considering the dust mass concentration c to be composed of a mean (time-smoothed) value $\bar{c}$ and a fluctuating component c' ,

$$c(x,y,t) = \bar{c} + c'$$

one derives the following time-smoothed equation for the diffusion of the solid phase in a two-dimensional, turbulent fluid flow (e.g., Rohsenow and Chou 1963):

$$\frac{\partial \bar{c}}{\partial t} + u \frac{\partial \bar{c}}{\partial x} + v \frac{\partial \bar{c}}{\partial y} = \frac{\partial}{\partial x} \left(D \frac{\partial \bar{c}}{\partial x} - \overline{u'c'} \right) + \frac{\partial}{\partial y} \left(D \frac{\partial \bar{c}}{\partial y} - \overline{v'c'} \right)$$

where

$\bar{c}$ = time-smoothed concentration

u = mean flow velocity in the x-direction

v = mean flow velocity in the y-direction

u' = turbulent velocity fluctuation in the x-direction

v' = turbulent velocity fluctuation in the y-direction

D = molecular diffusion coefficient.

The solution of the problem may be facilitated by means of the following assumptions: the expressions for the time-smoothed turbulent fluxes $\overline{u'c'}$ and $\overline{v'c'}$ are replaced by:

$$\overline{u'c'} = -k_x \frac{\partial \bar{c}}{\partial x}$$

$$\overline{v'c'} = -k_y \frac{\partial \bar{c}}{\partial y}$$

For a first approach the values of the eddy mass diffusivity, k_x and k_y , are constants; this would not be the case near a wall. After the passage of the shock wave, the mean flow velocity is uniform: $\bar{u} = U_\infty = \text{const}$, $v = 0$. Neglecting the molecular diffusion ($D = 0$) results in

$$\frac{\partial \bar{c}}{\partial t} + \bar{u} \frac{\partial \bar{c}}{\partial x} = k_x \frac{\partial^2 \bar{c}}{\partial x^2} + k_y \frac{\partial^2 \bar{c}}{\partial y^2}$$

Solving the above requires description of a set of initial and boundary conditions. The initial condition (index A) is given in the form of a system of sources which simulate the dust layer of length l and width b on the channel floor:

$$\begin{aligned} \bar{c}_A(x, y, t_1) &= 0 & \text{where} & \begin{cases} x < 0, y \\ x > 1, y \\ 0 \leq x \leq 1, y > 0 \end{cases} \\ \bar{c}_A(x, y, t_1) &= \infty & \text{where} & 0 \leq x \leq 1, y = 0 \end{aligned}$$

In each point of the interval $0 \leq x \leq 1$, the dust source is initiated by the shock wave that propagates with the velocity u_s and passes the leading edge of the dust layer ($x = 0$) at time $t = 0$. This initiation of the source activity is expressed by the second condition above; the instantaneous peak of the dust mass concentration at time t_1 can be described in terms of a Dirac function. The peak of dust concentration propagating across the interval $0 \leq x \leq 1$ represents a mass source of intensity $\phi(x, t_1)$ being active continuously during the time period $0 \leq t_1 \leq t$. The source intensity ϕ is the dust concentration $c(x, y, t)$ integrated over the total volume of the channel from $x = -\infty$ to $x = \infty$, yielding a finite value; this is the amount of dust mass m released by the source up to this instant of time t :

$$b \int_{-\infty}^{+\infty} \int_0^a c(x, y, t) dy dx = \int_0^1 \int_0^t \phi(x, t_1) dt_1 dx = m(t)$$

where b is the width (in z -direction) and a the height (in y -direction) of the shock tube channel.

Prediction of the development of dust mass concentrations in unsteady air flow is possible with the equation developed by Bracht (1979). However, it requires data such as initial flow conditions which can be determined only by means of experiments. (The original reference can be checked for the details of calculations using this model.)

2.3.2.3 Air Blast

Explosion air blast-entrained dusts were studied by Singer, Harris and Grumer (1976) in a 0.6 m dia by 50 m long explosion tunnel. Explosive generated airflows are characterized by wave-like responses: increasing flow followed by flow reversal. General findings were:

- Entrainment rates are a weak function of the instantaneous air speeds above the beds.
- Wetted, or wetted-dried, layers of coal dust on rock dust dispersed faster than untreated layers due to selective lifting of relatively large briquetted fragments.

Bulk quantities of dust were entrained simultaneously by longitudinal regression from the leading edge of the dust bed and by lifting of the surface layer. Figure 2.3-2 illustrates the successive stages of dispersal of cohesive-type dust beds. Dispersal began during the ascending gradient of the first transient airflow (and pressure) wave, and was usually completed by the end of the first quarter wave period.

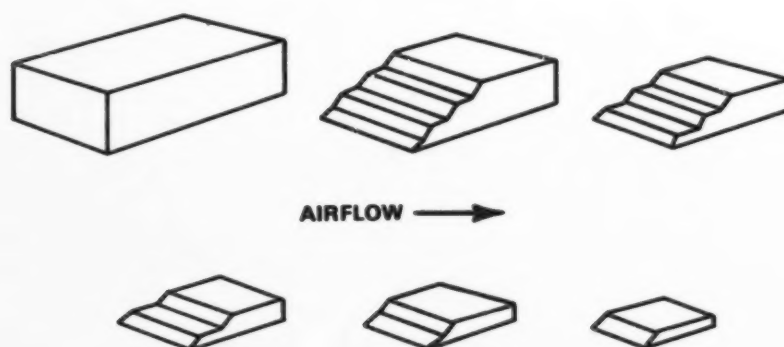


FIGURE 2.3-2. Diagram of Successive Stages of Dispersal of Cohesive-Type Dust Beds

Threshold instantaneous air velocities for entrainment of bulk quantities of coal and rock dust beds in separate, mixed and layered configurations were in the same range (5 to 30 m/sec) as those under steady-state conditions. Selective lifting of coal dust layers from rock dust occurred when the threshold dispersal velocity of coal dust (but not of underlying rock dust) was exceeded.

The empirically derived equations (Singer, Cook and Grumer 1972) were extended in this work and Froude numbers developed in two dimensionless components Fr_1 and Fr_2 :

$$Fr_s = Fr_1 \times Fr_2$$

$$Fr_s = \frac{\rho_F (U_*)^2}{(\rho_B S_c GH)^{0.5}} = \frac{\rho_F U_*}{(\rho_B S_c)^{0.5}} \cdot \frac{U_*}{(GH)^{0.5}}$$

where

Fr_s = Froude number, inertial force/restraining force (dimensionless)

ρ_F = bulk density of fluid, g/cm³

ρ_B = bulk density of powder, g/cm³

S_c = yield shear stress of bed material, dynes/cm²

U_* = friction velocity at threshold dispersal

G = gravitational constant

H = bed height.

The conservative forces are proportional to $(\rho_B S_c)^{0.5}$ in Fr_1 and to $(GH)^{0.5}$ in Fr_2 . U_* thus becomes proportional to $(\rho_B S_c)^{0.5}$ and $(GH)^{0.5}$, respectively, when Fr_s and either Fr_1 or Fr_2 are constant. Since $U_* \sim U^{0.88}$, then the critical velocity U_c is for threshold disposal proportional to $(\rho_B)^{0.57}$ and to $(\rho_B S_c)^{0.57}$, in fair agreement with two of the empirical equations for flat-topped cohesive coal and rock beds.

The experimental relationship $U_c = (H)^{0.52 \text{ to } 0.59}$ in Singer's earlier work also agrees with another theoretical concept based on a balance of drag and gravitational forces. The drag force F_D equals $C_D \rho_F (U)^2 \underline{A}/2$, and the gravitational force for spherical objects equals $G \rho_B (\pi H^2)/6$; $\underline{A}$ is the projected area equal to $\pi H^2/4$ for spherical objects, C_D is the drag force coefficient, and U is not defined. The critical balance of inertial force divided by restraining force is incorporated in Fr_c as follows:

$$Fr_c = C_D \rho_F (U_c)^2 \frac{(\pi H^2)}{4} \frac{G}{\rho_B} \frac{(\pi H^3)}{6}$$

Thus, $U_c = (H)^{0.5}$ when Fr_c is constant. The critical Fr_c ratio can be extended to include frictional f_* and cohesional S_c terms in the denominator:

$$U_c = (H)^{0.5} \left(\frac{\rho_B}{\rho_F} \right)^{0.5} (C_D)^{-0.5} (f_*)^{0.5}$$

The drag force coefficient C_D in the above equations depends on the roughness of the bed surface; C_D for single particles from a smooth plate is a function of Re_p , the Reynolds number of the particles. In streamline flow, $C_D = 24/Re_p$ for $Re_p < 1.0$; $C_D = 18.5/(Re_p)^{0.6}$ in transition flow for $1 < Re_p < 1,000$; and $C_D = 0.44$ in turbulent flow for Mach numbers less than 0.3 when $1,000 < Re_p < 200,000$. An expression for C_D proposed by Schlichting (1968) should also be applicable to the lifting of single particles or small aggregates from a stepped dust configuration by a turbulent field. According to Schlichting, in a turbulent stream along a roughened plate,

$$C_D = \left(2.87 + 1.58 \log \frac{Z}{K}\right)^{-2.5(a)}$$

where Z is the distance along the plate and K is the main height of surface protrusions. Along these lines, Chepil (1945) notes that entrainment of particles always begins with a rolling or sliding motion as the drag force exceeds the frictional force f . Punjraht and Heldman (1973) suggest that the sudden increased aerodynamic shear stress caused by transition from laminar to turbulent flow is a major factor in threshold movement of single particles from surfaces. Singer's experimental findings tend to agree with these observations.

When bulk aggregates or single particles are entrained from stepped dust beds instead of smooth surfaces, the characteristic bed height H is used in the present Fr_s ratio equations instead of particle diameter because clumps appear to be vertical segments of cohesive beds. The clump or aggregate size, which in all likelihood is itself a complex function of the flow velocity, the turbulence produced at the stepped edge, the yield strength of the bonds breaking under shear stress, and the sliding friction after separation cannot yet be quantitatively defined by these parameters. The substitution of H for the aggregate, clump, or particle size appears to be justified by the narrow range of critical Froude numbers, Fr_s , from 0.0077 to 0.038, calculated from

(a) $\log = \log_{10}$ in this report.

threshold dispersal data (Singer, Cook and Grumer 1972) for dust deposits of different geometry, height, bulk densities, and yield shear strengths.

2.3.2.4 Noncohesive Entrainment

A widely accepted equation for transport of granular noncohesive sediments (Shields 1936; Singer 1976) is:

$$\frac{\gamma_c}{(\rho_s - \rho_f)d_p} = f(Re_\star) = \beta$$

where

γ_c = critical shearing stress at the surface

ρ_s = solid density

ρ_f = fluid density

d_p = particle diameter

$Re_\star$ = shear Reynolds number $\left(\frac{U_\star \cdot d_p}{\gamma} \right)$

The minimum shear ratio β has been reported (Bogardi 1965) to be about 0.015 for intermediate Reynolds numbers $Re_\star = 10$. At these Reynolds numbers the surface grains are partially immersed in both turbulent and laminar boundary layers. The ratio increased to about 0.06 for $Re_\star \geq 400$ because of increased turbulence induced by bed roughness. Bagnold (1941) found $\beta = 0.01$ for incipient motion of particles in noncohesive granular beds by windflow and $\beta = 0.0064$ for saltation (the motion of particles with alternate contact with sand and air).

Threshold movement of particles of size d_p in saltating flow (Owens 1969, Singer, Harris and Grumer 1976) has been characterized with a Froude-type number, $Fr_{d_p} = \rho_f U_\star^2 / \rho_p g d_p$, with values in the range of 0.01 to 1.0. These Froude number values are about the same as those developed in Singer's dispersal data (1972).

2.3.2.5 Cohesive Entrainment

For cohesive dust beds, there are three distinct phases of dust movement to be accounted for:

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1. Incipient movement from the stepped leading edge of flat-topped beds
2. Initial slow entrainment of about the first 5 cm of the dust bed by regression from the leading edge and surface lifting
3. Accelerating entrainment of the remainder of the bed during accelerating flow.

The first two were considered by Bagnold (1941) for free-flowing material.

2.3.2.6 Free-Flowing and Cohesive Dust Entrainment Rates

Free-flowing and cohesive dusts have inherently different mechanisms of entrainment. Singer (1976) developed entrainment rate equations based on experimental data for explosion-induced dust entrainment (discussed earlier in this report:

free-flowing dust,

$$\bar{E}_{g/cm-width-sec} = 1.12 \rho_B^{10.9} H^{-0.47} \bar{U}_A^{2.2}$$

and cohesive dust,

$$\bar{E}_{g/cm-width-sec} = 1.06 \rho_B H^{0.72} \bar{U}_A^{0.47}$$

where

E = entrainment rate

$\bar{U}_A$ = average air velocity, m/sec

ρ_B = bulk density, g/cm³

H = height, cm.

Average entrainment rates of dust beds of complete bed lengths and ρ_B , $\bar{U}_A$, airflow acceleration $\Delta \bar{U}_A / \Delta t$ in m/sec², and pressure gradient $\Delta P / \Delta t$ in mbar/sec were correlated by regression analysis with the following results:

$$\bar{E}_{g/cm-width-sec} = 15.0 \rho_B^{0.93} H^{0.74} \bar{U}_A^{0.48}$$

$$\bar{E}_{g/cm-width-sec} = 7.5 \rho_B^{0.63} H^{0.72} \left(\frac{\Delta \bar{U}_A}{\Delta t} \right)^{0.39}$$

$$\bar{E}_{g/cm-width-sec} = 3.7 \rho_B^{1.25} H^{0.56} \left(\frac{\Delta P}{\Delta t} \right)^{0.43}$$

In these equations ΔU_A and ΔP are the differential increments of air velocity and pressure to the first pressure peaks, and Δt is the time interval from incipient particle movement to complete entrainment of the 20.5 cm beds. All $\bar{U}_A$ averages are based on graphical integration of areas under the velocity curves.

If the average air velocities U_A are transformed to friction velocities U_* , the Reynolds number change enables different dust sizes to be scaled into the equation. Thus, $(U_A)^{0.47}$ becomes $(U_*)^{0.6}$. A larger Reynolds number yields reduced values of friction factor f , friction velocity U_* , and wall shear stress τ (since $\tau = \rho_f U_*^2$), and lower entrainment rates result.

2.3.2.7 Surface Creep

Entrainment of stepped dust beds could be similar to hydraulic transport by surface creep of sediment at the bottom of a horizontal stream. Theoretical bed-load expressions have frequently equated $E_{g/cm-sec}$ to excess fluid shear stress τ , using either an incremental difference $\tau_o - \tau_c$, or a ratio τ_o / τ_c , where $\tau_o - \tau_c$ are available and critical fluid shear stresses at the surface of the deposit. One example Singer cited was:

$$E_{g/cm-width-sec} = A \tau_o (\tau_o - \tau_c)$$

where A = sediment parameter. The entrainment rate is proportional to $(\rho_f \bar{U}_*^2)^2$ and approximately proportional to $(\bar{U}_A)^{3.5}$. This is higher than the $(\bar{U}_A)^{0.5}$ Singer found for cohesive dusts.

2.3.2.8 Fluctuating and Steady Airflow

A different mechanism for particle entrainment was suggested by Einstein (1942), who believed entrainment did not depend essentially on a critical hydraulic quantity, but on fluctuations of air velocity at the surface. He proposed that surface particles migrate in distances that are proportional to the probability that flow drag on particles is sufficient to lift them into the stream. Surface velocity fluctuations rather than average velocity determine the probability. Other entrainment expressions were developed by

Kalinske (1947) and Graf and Acaroglu (1968). These assumed entrainment by statistically fluctuating pressure and velocity components. Kalinske's equation has the form

$$E_{g/cm-sec} = [(\rho_F d_p) U_*] [\phi(\tau_c/\tau_0)]$$

where ϕ is a function related to fluid turbulence, indicating a relatively weak dependence of E on U . Threshold shear velocity conditions were also defined by Graf and Acaroglu for several types of bed movement:

1. Starting motion, $\psi_{crit.} = 8 Re_*$ for $\psi_{crit.} \leq 20$
2. Formation of ripples and dunes for $\psi_{crit.} \leq 8$
 Beginning of ripple, $\psi_{crit.} = 0.4 Re_*$
 Disappearance of ripple, $\psi_{crit.} = 0.85 Re_*$
3. Bed-load movement and suspension for $\psi_{crit.} \leq 2.6$
 $\psi_{crit.} = 0.2 Re_*$
4. Sliding beds and suspension for $\psi_{crit.} \leq 1.4$
 $\psi_{crit.} = 0.06 Re_*$

where ψ is the shear intensity parameter, $f(\tau_c/\tau_0)$, $Re_* = U_* d_p \tau$, and U_* = velocity.

Partheniades and Passwell (1970) have developed an entrainment rate equation for erosion of cohesive sediments based on the concept that fluctuating flow components at the bed induce an instantaneous tensile stress within the bed that exceeds the weakest bond between flow aggregates. The weakest bond was considered to be more representative of the dispersing material than its macroscopic shear strength. Their entrainment equation reduces to

$$E = \frac{A' D_s \gamma_s}{t} [Pr]$$

where

A' = a shape factor

D_s = the average diameter of the entrained particles or aggregates

γ_s = the unit weight of each cluster

t = the time for breaking out of a clump from the bed

Pr = the probability that the instantaneous tensile strength in the deposit will exceed its cohesive bond strength.

Empirical entrainment rate equations that relate entrainment and air velocity may depend on experimental method. For example, Dawes (1952) developed the following entrainment equation, based partially on dimensional analysis and experiments:

$$E_{g/sec} = k U_* - (U_{*0})^3 \frac{\rho_B}{C}$$

where

k = a proportionality factor

U_{*0} = the friction velocity at threshold velocity

ρ_B = bulk density

C = a cohesive property of the dust bed.

Bagnold found that entrainment rates for sand movement over a flat plate followed a U_*^3 dependency, based on weight loss during incomplete dispersal in steady flow. Hsu (1971) reported the rate of eolian sand transport to be proportional to $[U_*/(gD)^{1/2}]^3$, where D is the grain diameter.

Investigations have all shown agreement on the dependence of the entrainment of free-flowing dusts on air velocity in steady airflow:

$E \approx U_A^2$ or U_A^3 . Cohesive dust entrainment rates were found by Singer, Harris and Grumer (1976) to be related to $U_A^{0.3}$ to $U_A^{0.5}$, based on complete dispersal of the dust bed. This agrees with theoretical work where dust bed entrainment was governed by fluctuating and drag components, thus inducing an instantaneous stress within the bed exceeding the weakest bond between dust aggregates.

Singer indicates that the computed entrainment rates can be within a factor of 2 to 4 of experimental values, as shown in Table 2.3-8.

TABLE 2.3-8. Entrainment of Dust Beds Produced by Air Generated in an Explosion

Coal-Dust Bed Dimensions, cm			Air Velocity U_A , m/sec	$\Delta P/\Delta t$, atm/sec (b)	Entrainment Rates (a)		
					E_{pred}		E_{exp}
Length	Width	Height			5.0-cm bed (c)	20.3-cm bed (d)	
89	104	7.6	18.9	0.6	0.6	2.2	1.1
89	104	7.6	53.7	0.47	0.5	1.8	0.4
89 ^(e)	104	7.6	12.4	1.68	0.7	2.5	0.5
150	15	5.1	112.5	1.67	2.5	8.6	3.8
150	15	2.5	112.5	1.67	1.1	3.9	1.3
150	15	1.9	112.5	1.67	0.9	3.1	1.0
150	15	7.6	100	1.55	3.2	11.6	4.5
150	15	2.5	100	1.55	1.1	4.0	1.0
150	15	2.2	100	1.55	0.8	2.6	0.6

(a) g/cm width/sec

(b) $\Delta P/\Delta t$ is the average gradient to the first peak of the pressure wave, using the sonic velocity approximation $P = \rho F C U_A$.

(c) Based on wind tunnel experiments, $E_{pred} = 1.06 \rho_B H^{0.72} U^{0.47}$ for 5.0 cm bed length.

(d) Based on explosion tunnel experiments, $E_{pred} = 15 \rho_B H^{0.74} U^{0.48}$ for 20.3 cm bed length.

(e) 0.64 cm coal dust layer on 6.96 cm thick mixture of 80 percent rock dust and 20 percent coal dust.

2.3.3 Entrainment from Spills

Aerodynamic entrainment can result from powders falling through a flowing airstream. Sutter (1980) performed experiments pumping fluidized soil into the top of a moving airstream within a wind tunnel 2 ft high, 2 ft wide and 30 ft long, and developed a regression equation relating the fraction of source airborne to the wind speed:

$$y = 0.00599x + 0.00543$$

where

y = fraction of source airborne

x = wind speed, mph.

Entrainment releases measured in these experiments were considered the maximum for entrainment mechanisms, since the soil was immediately airborne.

2.3.4 Relative Humidity Effects

Relative humidity could affect the adhesion of particles to a surface. At elevated humidities, particles can pick up moisture from the air and totally or partially dissolve, forming a very strong bond with the surface. Hygroscopic properties of particles appear to be a major factor in the adhesion of particles to a surface. Most powders in nuclear fuel cycle facilities are not hygroscopic but RH could have some effect on pickup. Spilled solutions might be more responsive to RH. "Percent Particles Remaining After Blowoff" was designated "Retention Percent" in the reported experiments:

$$\text{Retention percent} = \frac{\text{count after blowoff}}{\text{count before blowoff}} \times 100.$$

Figure 2.3-3 shows a comparison plot of total particles retained for all RH treatment levels. It can be seen that RH is important, with particles in a humid environment more likely to be retained on the surface.

The effects of RH are shown by Whitfield (1978), who performed experiments using 20 psi nitrogen blowoff as a removal mechanism for ambient dust from <1 μm up to 140 μm . Slides were conditioned at different humidities (33 percent to 100 percent) in a humidity control chamber, removed and subjected to

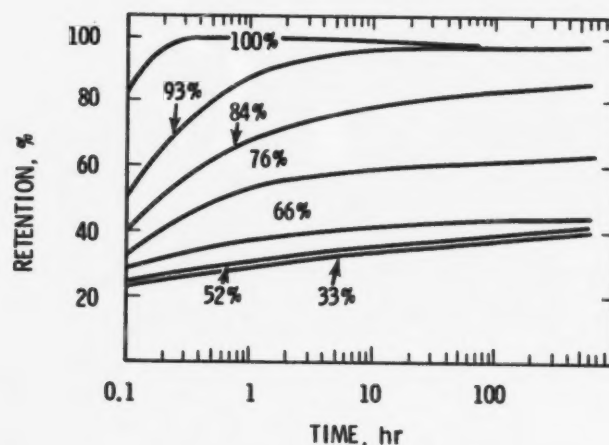


FIGURE 2.3-3. Effects of Relative Humidity on Surface Particle Retention

the blowoff for 10 sec. Temperatures during the tests were not indicated. Particle adhesion increased drastically as RH increased above 50 percent, with the greatest changes in the first hour of conditioning time.

2.3.5 Agglomerate Breakup

Airborne agglomerates can break up after aerodynamic entrainment. Caveny (1979) developed a correlation of the minimum agglomerate size for breakup of Al/Al_2O_3 . This was obtained in terms of the ratio of Al/Al_2O_3 surface tension and shear forces. Generally, agglomerate breakup occurs when the Weber number (ratio of inertial forces to surface tension forces) exceeds the range of 20 to 30:

$$We = \frac{d_{ag} \rho_g (U_g - U_{ag})^2}{\sigma}$$

where

We = Weber number

d_{ag} = diameter of agglomerate, μm

ρ_p = density, kg/m^3

U = velocity
g = gas
ag = agglomerate
 σ = surface tension, N/m.

This experimental work shows the influence of gas flow on agglomerated matter, where agglomerates are subjected to shear conditions that influence breakup. As the flow accelerates, the velocity differential between the agglomerates and the gas deforms the agglomerates, causing them to break into small droplets.

The breakup time of a droplet suddenly exposed to a gas stream has been shown (Harrje 1972) as

$$t_b \approx \frac{d_{ag}}{2(U_g - U_{ag})} \left(\frac{\rho_{ag}}{\rho_g} \right)^{1/2}$$

2.3.6 Motion of Sand

Bagnold (1941) stated that our knowledge of the fundamental relations between particle size and movement and fluid velocity is scanty and does not rest on a theoretical basis. His studies are reported in the classic work, Blown Sand and Desert Dunes. His work was concerned with the movement of solid noncohesive particles and wind--differing from ours, which studies fine powders and locations within structures. He classified three possible types of grain motion:

- Suspension--particle never comes in contact with ground
- Surface Creep--particle always remains on ground
- Saltation--alternate contact with air and ground.

Particles entering local winds over a storage area as a result of wind erosion were studied by Davies (1980). He described the three different mechanisms of Bagnold, using some work formulated by earlier investigators.

The critical horizontal drag force F_c for spherical particles was given by Chepil and Woodruff (1963) in the form

$$F_c = \left(\frac{\pi}{6} g_D^3 \rho' - L_c \right) \tan \phi$$

where

g = acceleration due to by gravity

D = equivalent diameter of the material

ρ' = mass density of the material

L_c = aerodynamic lift force

ϕ = angle of repose.

Furthermore, Chepil found that $L_c \approx 0.85 F_c$ when erosion conditions existed. Hence the drag force on the particles takes the revised form

$$F_c = \frac{\frac{\pi}{6} g_D^3 \rho' \tan \phi}{1 + 0.85 \tan \phi}$$

By dividing this expression by the cross-sectional horizontal area of the particle, $\pi D^2/4$, the threshold drag F_c per unit cross-sectional horizontal area occupied by a grain is obtained:

$$F'_c = \frac{0.66 g \rho'_D \tan \phi}{1 + 0.85 \tan \phi}$$

To generalize this expression for turbulent wind flow over a surface two factors were introduced, η and T ; η reflects the ratio of drag and lift on the upper exposed particles to the total drag and lift of the surface, and T is the ratio of peak wind pressure to mean wind pressure. Thus the equation can be restated as

$$\tau = \frac{0.66 g_D \rho' \eta \tan \phi}{1 + 0.85 \tan \phi T}$$

which is the critical surface shear stress above which particles will become mobile. The wind induced surface shear stress is given by

$$\tau = \rho U_*^2$$

where ρ is the mass density of air and U_* is the friction velocity. The measurement of U_* involves obtaining detailed information on the turbulence structure at a site and is an inconvenient estimator. Alternatively this value can be obtained from the "law-of-the-wall" expression

$$\frac{U}{U_*} = 2.5 \ln\left(\frac{z}{z_0}\right)$$

where U is the mean wind velocity at height z and z_0 is the height where $U = 0$ and is related to the height of the roughness elements of the surface.

Sutton (1953) provided values of z_0 for various surfaces:

Very smooth surfaces (mud flats, ice)	$z_0 = 0.001$ cm
Lawn grass up to 1 cm high	$z_0 = 0.1$ cm
Thick grass 10 cm high	$z_0 = 0.7$ cm

where z_0 is effectively independent of wind speed. It has been shown (Davenport 1968) that the circular Weibull distribution provides a good fit to the wind speed data and describes the probability of exceeding a given wind speed from a given direction:

$$P(>U, \theta) = A(\theta) \exp \left[- \frac{U}{C(\theta)} \right]^{K(\theta)}$$

where $A(\theta)$ is total frequency of occurrence of winds from the direction that will yield $\sum A(\theta) = 1.0$, and $C(\theta)$ and $K(\theta)$ are curve-fitting coefficients. Hence the probability of exceeding a critical surface shear stress is given by

$$P(>U_{*}^C \theta) = A(\theta) \exp - \left[\frac{U}{C(\theta)} \right]^{K(\theta)}$$

where U_{*}^C is the critical value of the friction velocity and U , the mean wind velocity, is given by the equation $(U/U_{*}) = 2.5 \ln(z/z_0)$. Summing the probabilities for each direction of exceeding critical friction velocity, i.e.,

$$\sum_{\theta} P(>U_{*}^C, \theta)$$

gives the frequency of occurrence as a proportion of time that wind erosion will be evident at a site.

In general this procedure provides a coarse estimation of the field conditions since many other factors such as compaction, binding agents, moisture content, freezing, thawing, and precipitation all modify the surface descriptors z_0 and n , which in turn affect the threshold surface shear stress.

Therefore, in order to circumvent the above and acquire an engineering estimate of the frequency of erosion conditions, wind tunnel tests can be undertaken on preconditioned samples of the material to directly determine the critical mean wind speed U_c , which will induce particle movement at a given height. The equation

$$P(>U_{*}^C, \theta) = A(\theta) \exp \left[- \frac{U}{C(\theta)} \right]^{K(\theta)}$$

can then be used directly.

The analysis to this point pertains to winds shearing on a plane with the winds unaffected by stockpiles and other obstructions. The measurements of wind speed undertaken on the model are normalized by the reference wind speed U_{ref} corresponding to the location of the nearest meteorological station. These ratios

$$R_i(\theta) = U_i/U_{ref}$$

where i defines a specific point on the model, range from 0 in calm areas to >2 in areas which undergo accelerations due to the site characteristics. These local increases in velocity imply that the approaching winds can have relatively low speeds and still induce significant particle movement. The equation modified to include $R_i(\theta)$ is

$$P_i(>U_c, \theta) = A(\theta) \exp - \left[\frac{U_c}{R_i(\theta)C(\theta)} \right] K(\theta)$$

This provides an engineering approximation of the relative frequency of wind erosion over the irregular surfaces found at most storage facilities.

2.3.7 Entrainment of Volcanic Ash

One final example of aerodynamic entrainment operating to resuspend freshly deposited materials was dramatically illustrated after the May 18, 1980 eruption of Mt. St. Helens in Washington state. Airborne volcanic ash levels reached $35,000 \mu\text{g}/\text{m}^3$ in Spokane on the day the mountain erupted and this subsequently settled to the ground (Tri-City Herald 1980). On June 2, 1980, high winds entrained the ash, and particulate levels soared to $1000 \mu\text{g}/\text{m}^3$, 12 times the usual level. As can be seen, the aerodynamically entrained particulate level was below that sustained in the initial event by a factor of 35, but still was significantly above background. In accident analysis, this phenomena is one of the incremental portions that make up the total release. If it extends for a significant time period, much or all of the settled material can become airborne.

2.3.8 Summary

Several approaches to estimating airborne releases from aerodynamic entrainment have been reviewed. As credible nuclear fuel cycle accidents are scoped, they should be analyzed to determine which method is appropriate to use. To illustrate, Mishima's work was designed to simulate fuel cycle accidents, using suitable surrogates and situations for these facilities.

Singer's studies investigated entrainment produced by high-energy explosions of coal dust, producing a short (msec) duration deflagration. Although deflagrations might be important potential fuel cycle accidents, coal dust is not a suitable surrogate.

Much of the work reviewed developed empirical relationships for aerodynamic entrainment. These methods must be used judiciously and thought given to their applicability to the specific accident scenario.

2.4 EXPLOSIONS AND PRESSURIZED RELEASES

The term "explosion" describes a process in which the rapid production or release of gas under high pressure causes a pressure wave (blast) to propagate through the surrounding medium, which is usually air (Strehlow 1980). The blast is called a free-field blast until it interacts with objects in its path. This section will consider any pressurized release of contained material including those forcibly ejected at less than blast pressure.

Accidental explosions can cover a wide spectrum of events, which have been grouped into nine major categories, each with its own distinctive characteristics. Table 2.4-1 shows this categorization of explosions and where they are likely to occur. This listing covers a wide range of severity, not all applicable to the nuclear facilities we are considering or to a lesser than maximum degree of intensity. They are, however, events we have investigated to help us understand explosions.

The most common types of explosions conceivable in a MOX facility (Selby 1975) are 1) hydrogen explosions in sintering furnace, 2) autoclave explosions, and 3) chemical explosions. Other potential explosions are resin column and glovebox explosions, which are highly localized. No explosions could be conceived of that would involve the entire facility and result in major damage.

A generic environmental impact statement (USAEC 1974) indicated potential explosions could occur at the sintering furnace, clean scrap recovery operation, dirty recycle operation, or at locations where combustible material may be located. All postulated releases were estimated to be the same as those in a fire.

TABLE 2.4-1. Categories of Accidental Explosions and Where They Are Most Likely to Occur (Strehlow 1980)

Type of Explosion	Typical Situations
Condensed phase detonations	Chemical reactors, distillation columns, separators, factories that produce high explosives and propellants
Combustion explosions of gaseous or liquid fuels in enclosures	Buildings, ships, tankers, boilers, compressed-air lines
Combustion explosions of dusts in enclosures	Coal mines, grain elevators, pharmaceutical industry
Boiling-liquid, expanding-vapor explosions (BLEVEs)	Ductile vessels such as tank cars containing combustible, high vapor-pressure liquids
Unconfined vapor-cloud explosions	Spills of highly volatile fuels, chemical plants
Explosions of pressurized vessels containing nonreactive gaseous materials	Boilers, pressurized gas tanks
Explosions resulting from chemical reactor runaway	Chemical reactors
Physical vapor explosions	Island volcanoes, molten material poured into a moist container
Explosions resulting from nuclear reactor runaway	Nuclear reactors

In the chemical industry, combustion explosions and the handling of explosive liquids account for more than 75 percent of the explosion losses (Doyle 1969). This study classified the explosions into the types shown in Table 2.4-2.

The discussion above generally covers the pressurized releases most likely to occur in a nuclear fuel cycle facility. Other more extreme events, such as nuclear explosions, have been investigated to gain an understanding of the phenomenon. Accidental explosions can result in detonation, ignition, and combustion, some producing a fireball.

TABLE 2.4-2. Types of Explosions in a Chemical Industry Study, 1964 to 1968

<u>Type of Explosion</u>	<u>Number of Explosions</u>
<u>Combustion</u>	
In equipment	13
Outside equipment in building	8
In open	<u>1</u>
TOTAL	22
<u>Reaction</u>	
Explosive liquid or solid	12
Runaway reaction	<u>4</u>
TOTAL	16
<u>Metal Failure</u>	
Corrosion	1
Overheating	3
Accidental overpressure	<u>2</u>
TOTAL	6

In discussing explosive release, the term TNT equivalence is frequently used. The standard for TNT energy release is 5×10^5 cal/lb (Grelecki 1974). Table 2.4-3 shows the equivalent TNT values for several pressures, which can be used to relate explosive releases. These equivalents assume that a catastrophic breach is all the material leaving at once.

This report is concerned with accident-generated aerosols: the quantity that becomes airborne and the airborne particle size distribution of that mass. In trying to find this information, all kinds of explosions were

TABLE 2.4-3. TNT Pressure Equivalents

<u>Pressure, psig</u>	<u>TNT Equivalent, lb/ft³</u>
10	0.001
100	0.02
1000	0.42
10000	6.53

studied extensively and calculations were developed to estimate the effects of shock waves, explosibility of powders and dust, temperature rise, pressure rise, maximum explosion pressure, etc. (Stull 1977; Strehlow 1975; Jacobsen et al. 1964; Dorsett 1968; Nagy 1968; White 1946). The information directly useful for estimating airborne particulate releases from accidental explosions involving PuO_2 and mixed oxide powders is limited.

Other investigators at Argonne National Laboratory (Seefeldt and Mechem 1977) have done an extensive literature search in the field of explosive releases specifically directed toward fuel reprocessing and related fields. This information will be included liberally in our review. They could find no experimental work that would follow the release history of aerosol and dust clouds immediately following a event.

2.4.1 Dust Explosions

Powders involved in an industrial process can explode; fine dusts in a divided airborne state are a well-known explosive hazard. Hartman (1943) showed that particle size and explosibility of metal dusts are related: as the smaller particle concentration in the dust increases, the minimum explosive concentration is lowered, as illustrated in Table 2.4-4.

An empirical explosibility index has been developed that is related to ignition sensitivity and explosion severity. Uranium dust, with an average particle diameter of 10 μm , has an explosibility index >10 (Jacobsen 1964). The minimum explosive uranium dust concentration in air is 0.06 oz/ft³

TABLE 2.4-4. Relationship Between Dust Fines and Explosive Concentrations

Fines Fraction, Percent (a)	Minimum Explosive Concentration, g/
1	0.125
15	0.060
40	0.040
92	0.025

(a) <200 mesh, 76 μm .

(60 g/m³) of dust dispersed. Because of careful controls, this level of uranium airborne is unlikely in fuel cycle facilities, so a dust powder explosion is not a potential accident.

Combustible quantities of airborne dust normally exist inside ducting or process equipment only (Strehlow 1980). A small explosion in such equipment causes it to rupture and throw burning dust into the workplace. If the work area is dirty, the resulting gas motion and vibration of equipment could cause the layered dust to become airborne and fuel more explosions. Dust explosiveness has not been reviewed as a serious problem for MOX facilities.

2.4.2 Industrial Process Releases

Studying pressurized industrial processes could give insight into potential releases in a nuclear facility. For example, a plutonium oxalate slurry could be in a spray that is explosively jetted into a room. This could be compared to ceramic spraying or metal deposition spraying, both of which have been studied (Danielson 1973). Air pressures used for ceramic spraying can range from nearly 0 to 125 psig. The particulate matter in the unfiltered exhaust approaches a concentration of 5 grains/scf (12 g/m³) for heavy spraying; up to 10 grains/scf (23 g/m³) has been observed. These airborne particles have a high affinity for water, strong agglomerating tendencies when wet, and relatively high unit weight; thus they are easily removed from the air.

Higher spray pressures result in smaller droplet sizes, which can be predicted by the equation:

$$D_o = \frac{1920}{Y} \left(\frac{\sigma}{\rho} \right)^{0.5} + 597 \left(\frac{\mu}{\sqrt{\sigma \rho}} \right)^{0.45} \left(\frac{1000 Q_1}{Q_2} \right)^{1.5}$$

where

D_o = mean drop diameter, μm

Y = relative velocity of the gas with respect to the liquid, fps

σ = surface tension, dyne/cm

ρ = fluid density, g/cm³

μ = fluid viscosity, poises

Q_1/Q_2 = ratio of liquid volume to gas volume.

Since Q_1/Q_2 is usually very small, D_0 is determined primarily by the first term. Since $Y \approx \sqrt{P_s}$ and σ and ρ are essentially constant for the same type of operation, the equation can be simplified:

$$D_0 = \frac{k}{\sqrt{P_s}}$$

where

D_0 = mean drop diameter (defined as a drop with the same ratio of volume to surface as the total sum of all drops formed), μm

k = constant, empirically determined

P_s = atomization pressure, psig.

Values of k were ≤ 1200 in ceramic spraying operations, depending on the number of baffles in a system. Droplet size decreases because larger droplets are removed by each succeeding stage.

2.4.3 Nuclear Explosions

Information generated from nuclear explosions was reviewed for particle properties that could result from other pressurized release events. For example, a concise summary of the physical and radiochemical properties of particles observed in nuclear debris fallout was compiled by Crocker (1966). These types of bursts were identified:

- high air--no soil involved
- low air and tower--sometimes involving soil
- ground surface--involving large amounts of soil.

The properties of the airborne particle are listed in Table 2.4-5. The releases were primarily oxides of uranium, plutonium, and structural metals. The values in the table permit valid comparisons with radioactive debris particles from other sources. There was little information on submicron particles.

TABLE 2.4-5. Properties of Fallout Particles

Variables	High Air Burst	Low Air and Tower Bursts	Ground Surface Bursts
Size	Seldom greater than 20 μm diameter. Distribution unknown. Size varies inversely with yield.	Range from a few microns to several hundred or a few thousand microns. Upper limit depends upon nature and amount of interacting soil, etc.	Same as low air and tower bursts.
Shape	Almost all spherical. Occasionally two spheres are stuck together.	Both spheroidal and irregular. Proportions depend on shot conditions. Small spheres stuck to larger particles sometimes reported.	Irregular particles predominate. Some spheroids observed. Various kinds of agglomerates and partially melted particles.
Particle density	3-4.3 g/cm^3	1-3 g/cm^3	1-3 g/cm^3
Fractionation	Larger particles ($>1 \mu\text{m}$ dia) depleted in volatiles behaving mass chains. Submicron particles relatively representative.	Close-in fallout is usually depleted in volatiles behaving mass chains, while world-wide fallout is enriched.	Cratering shots may trap large amounts of refractory mass chains in fallback. In these cases, local fallout is enriched in volatiles behaving mass chains.

These comparisons indicated higher density particles when no soil is involved; particles are larger when there is soil interaction. Depending on the characteristics of the device and the shot conditions, particle sizes may range from submicrons to centimeters. The activity may be concentrated on the particle surface or uniformly distributed throughout the particle.

Particles generated by Project Buggy at the Nevada test site (Cohen 1970) were sized using cascade impactors and analyzed for gross beta emitters. Nine

and one-half percent of the particles were $\leq 10 \mu\text{m}$ in the samples taken about 15 min after the event. (The results in Table 6, p. 8 of that report were plotted but not included here.) A second sizing at 55 min after the explosion showed the diameters were an order of magnitude greater than in the first sample. This was thought to result from deeper penetration of the samplers into the plume. After one hour there was a lower standard deviation among samples, indicating a more uniform cloud concentration. The radioactive material was found in the leading edge of the cloud.

Gamma counting gave no indication of the mass. Cohen ignored the fraction greater than $25.3 \mu\text{m}$ and calculated a mass median diameter (MMD) of $4.5 \mu\text{m}$ and σ_g of 2.5.

The characteristics of the airborne material from Project Schooner (Gudiksen 1970) included the particle size distribution and mass concentration as a function of time, as shown in Table 2.4-6. The MMD became smaller and the mass concentration airborne decreased as a function of time after an event, as might be anticipated.

Particle size distributions of the aerosols produced in Operation Roller Coaster (Friend 1965) were determined by cascade impactor collection. The AED was $40 \mu\text{m}$, σ_g was 4, with 7 percent of the particles $\leq 10 \mu\text{m}$.

Seefeldt (1977) indicated some difficulties in using the data from nuclear explosions:

- The magnitude of the event is much larger than would be anticipated in a nuclear facility.

TABLE 2.4-6. Characteristics of Airborne Material from Project Schooner

<u>Time After Event, min</u>	<u>Mass Mean Diameter, μm</u>	<u>Geometric Standard Deviation</u>	<u>Maximum Mass Concentration, Observed, mg/m^3</u>
12	30	2.8	240
40	20	2.8	100
70	17.5	2.6	50

- The explosion was unconfined; in a plant, walls and barriers would have an influence on how the materials would become airborne.
- The nature of the materials involved is different.

2.4.4 Fuel Cycle Explosions

In a review article Seefeldt (1976) summarized explosion source term data estimated in environmental reports for nuclear fuel cycle facilities. These are summarized in Table 2.4-7.

TABLE 2.4-7. Summary of Explosion Source Term Data from Environmental Reports

<u>Operation</u>	<u>Pu Released, μCi</u>
Fabrication of MOX Fuel ^(a)	7.0×10^1
Reprocessing ^(b)	2.5×10^4
Fuel Fabrication ^(b)	2.9×10^1

(a) Generic environmental impact statement for mixed oxide.

(b) LMFBR environmental impact statement.

2.4.5 Release Incidents

Seefeldt (1976) reviewed the size of airborne plutonium particles at nuclear facilities. These are listed in Table 2.4-8. All of the median diameters are in the respirable size range. He also compiled information on explosive accidents experienced in working with plutonium; the data are shown in Table 2.4-9. In all of the incidents Seefeldt listed, the plutonium release from the containment was very low. Even in the case of an accident at Oak Ridge National Laboratory (ORNL), in which 150 g of an aerosol of plutonium (probably very finely divided) was dispersed explosively into a cell, only 0.6 g escaped through an open cell door so there was only minor spread of plutonium. The information as presented does not seem to lend itself to back-calculation of source terms or release factors.

TABLE 2.4-8. Mean Plutonium Size Characteristics of Aerosols at Five AEC^(a) Facilities Upstream from Air-Cleaning Facilities

Location	Particle Diameter, μm	Geometric Standard Deviation, σ_g	Range of Particle Sizes, μm ^(b)
00	1.8	2.1	0.4 to 8.7
04	2.3	3.5	0.2 to 28
0.8	4.0	1.7	1.4 to 12
11	0.34	5.4	0.01 to 9.9
14	2.7	2.4	0.5 to 16

(a) Atomic Energy Commission

(b) Based on $2 \sigma_g$ for log-normal distribution. Lower and upper limits are obtained by respectively dividing and multiplying the mean by $2 \sigma_g$.

Operational nuclear incident data were compiled by Mishima (1969). One of these incidents, a plutonium oxide powder leak with helium gas, might be useful in estimating source terms. This pressurized leak released 300 mg of a 1.5 g source into a room, which yielded a release factor of 2×10^{-1} , and it must be assumed this was deposited in the room.

A chemical explosion in 1976 of an ion exchange resin column and the resulting americium contamination can be back-calculated to estimate a fractional release (ERDA 1976). In this explosion a maximum of 1 mCi ^{241}Am was released after an explosive vessel failure, with an explosive energy yield of 59 kcal (100 psi). Stack filters collected 50 μCi . The column inventory was 175 g, so at a specific activity of 3.45 Ci/g, the total inventory was 604 Ci. The fractional atmospheric release could be on the order of 10^{-6} to 10^{-7} because of estimation errors.

In another incident, heat buildup in a plutonium oxide storage can (Committee Report 1979) along with the presence of decomposition products and water in the plutonium oxide, led to the pressurization and subsequent rupture of the can when it was removed from a shipping container. There was a visible cloud of PuO_2 ejected from the container, which ruptured at an estimated 60 psi. Approximately 1.2 to 1.3 mCi escaped in less than an hour, with maximum

TABLE 2.4-9. Major Plutonium Explosions

Date	Damage Category	Location	Manner and Amount of Pu Release	Explanation
11/16/51	Criticality excursion in criticality test	Richland, Washington	Pressure buildup inside a vessel containing Pu solution caused the solution to spray into room. No environmental release.	Too-rapid withdrawal of a safety rod resulted in criticality and boiling surge. No ventilation system damage.
3/31/59	Chemical explosion	Pu Metal Machining, Richland, Wash.	No significant contamination outside the laboratory room; glove box breached, room contamination up to 26,000 dis/(min)(cm ²).	Explosion, apparently from air and possibly an organic solvent from sparks from Pu metal machining.
11/59	Explosion	Oak Ridge National Laboratory, Tennessee	150 g Pu in aqueous solution was ejected from the equipment into the cell. 0.6 g was blown through the cell door. Outside local contamination was extensive. Cell ventilation filters removed 1.5 g and did not allow release via this path to the plant stack.	A chemical explosion occurred in an evaporator inside a shielded cell.
5/11/61	Over-pressurized container	Miamisburg, Ohio	Room contaminated by leak through glove box vent. No environmental contamination.	A leak in a vacuum connection to a scrubber for the glove box operation resulted in the scrubber venting to the room. A ²³⁸ Pu precipitate was being dried in the glove box.
10/27/64	Chemical explosion	Miamisburg, Ohio	Pu escaped from the glove box, and extensive contamination of the building resulted from the spread of ~0.5 g of ²³⁸ Pu.	17 g of ²³⁸ Pu was being processed in a glove box having a methanol-air atmosphere. It is believed that a hot plate ignited the atmosphere and led to an explosion which pressurized the glove box and tore six gloves from the glove ports.
1/20/65	Over-pressurized container	Livermore, California	Some Pu discharged through the stack. No contamination was detected outside the building.	Pu-contaminated gas was released to a ventilated hood and was discharged to the atmosphere through a high stack.

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ventilation exhaust air concentrations reaching $1.4 \times 10^{-10} \mu\text{Ci}/\text{cm}^3$. The half-life was used to calculate the specific activity for each of the isotopes in the PuO_2 and this in turn was used to calculate the activity/g. These values are listed in Table 2.4-10. The ruptured can contained 1300 g PuO_2 ; therefore, at 9.36 Ci/g the total inventory was 12,168 Ci. Using the 0.0013 Ci release value, a fractional release of about 1×10^{-7} was calculated. Using the PuO_2 specific activity and the $\mu\text{Ci}/\text{cm}^3$ of the airborne PuO_2 , total mass in the exhaust was $1.4 \times 10^{-11} \text{ g}/\text{m}^3$. Insufficient description of the room prevented us from estimating the room concentration.

TABLE 2.4-10. Isotopic Composition of PuO_2 Involved in Pressurized Container Rupture

Isotope	Percent	Half-Life, y	Specific Activity, Ci/g	Activity per g(a) of PuO_2 , Ci
^{238}Pu	1.49	87.74	1.7×10^1	0.022
^{239}Pu	59.50	24,110	6.2×10^{-2}	0.032
^{240}Pu	23.98	6,537	2.3×10^{-1}	0.048
^{241}Pu	10.33	14.4	1.0×10^2	9.27
^{242}Pu	4.70	3.76×10^5	4.0×10^{-3}	0.0002
TOTAL	100.00			TOTAL 9.36

(a) Calculated assuming the PuO_2 is 87 percent Pu, since the formula is a symbolic representation of plutonium oxide.

2.4.6 Coal Mine Explosions

Coal mine explosions have been studied extensively, with some emphasis on dispersal of debris, since this dispersed debris can, in turn, initiate secondary explosions. Coal dust explosions can be directly related to volatile coal content (Grumer 1974). Volatiles could not be considered as important in powder explosions in a fuel cycle facility, so coal explosion is not directly comparable to dusts with lesser volatility. In fact, rock dust has been used to control coal dust explosions.

Dust movement in a coal mine explosion is consistent with known dust behavior; i.e., when dust is carried in the air currents in a mine, coarse dust particles settle rapidly to the floor close to the point where they are formed. Fine particles remain airborne and are often transported for relatively long distances.

Mass concentrations of dispersed material for coal ranging in size from 100 to 150 μm measured in a wind tunnel were related to air velocity and bulk density (Singer 1972). This coal was called free-flowing because of its large particle size.

This relationship is:

$$E = 31.2 U_L^{1.1} \rho_B^{5.9}$$

where

E = mass concentration, g/m^3

U_L = air velocity at mid-height of the compacted coal, m/sec

ρ_B = bulk density of the compacted coal, g/cm^3 .

The mass concentrations for air velocities of 5 and 40 m/sec are about 10 and 90 g/m^3 , respectively, for the bulk coal density of 0.604 g/cm^3 . The 5 m/sec value of 10 g/m^3 is two orders of magnitude larger than the 0.1 g/m^3 value that Sutter (1980) calculated for soil already normally entrained in a wind tunnel. This equation appears to be useful for coal only; using an estimated depleted uranium bulk density of 2.25 g/cm^3 in the equation yielded an airborne concentration about $2 \times 10^4 \text{ g/m}^3$. It is unreasonable to expect more of the heavier material to become airborne.

Later dispersal tests (Singer 1976) showed little apparent relationship between air velocity and solids dispersal. Mass concentrations of coal-rock mixtures ranged from 4 to 30 g/m^3 for air velocities of 9 to 65 m/sec . Comparable numbers for rock only were 10 to 60 g/m^3 for air velocities of 11 to 70 m/sec . Data indicate airborne material concentrations up to several hundred grams/m^3 can be obtained.

2.4.7 High Explosive Releases

Some work has measured airborne material produced from other explosive events. Project Middle Gust (Freeman 1972) was a non-nuclear event in which a 100-Mg sphere of TNT was exploded. Maximum mass concentrations from this event ranged from 700 to 1100 g/m³ and occurred within 1 sec of the detonation. A rapid decrease in concentration with time was indicated. Similar information was obtained using high explosives (Banister 1955). Blast-induced debris collected on greased stakes and specially designed collector plates measured dust concentrations ranging from 300 to 2400 g/m³.

In cratering experiments Miller (1962) inferred the distribution of large solid airborne particles from particle size analyses of materials deposited within a short distance of an event. At 105 m from the event, the mean particle size was 2200 μ m; at 335 m it was 170 μ m. The range of size distributions expressed by the geometric standard deviation was extremely wide, $\sigma_g = 11$, at the nearer location, and $\sigma_g = 2$ at the more distant locations. This indicates a smaller, more uniform size distribution farther from the source.

2.4.8 Natural Phenomena

The volcanic eruptions of Mt. St. Helens (see Section 2.3) have provided the opportunity to collect data on explosive release of material. Preliminary data from May 22, 1980, the first four days after the first eruption, found a total mass concentration of 11 μ g/m³ at 66,000 ft, about an order of magnitude above the ambient concentration for this region (Rawls 1980). The particles ranged in size from 1.6 μ m to <0.05 μ m. Five days later the concentration of airborne particles at 55,000 ft was about 6.5 μ g/m³.

2.4.9 Other Data

Swain and Haberman (1961) reviewed data from non-nuclear sources and calculated that 33 mg/m³ was the maximum airborne concentration a few minutes after an accident. Castleman, Horn and Lindauer (1969) generated fine particles using an exploding wire technique and found concentrations as high as 71 mg/m³ plutonium a few minutes after generation. Mishima (Selby 1975) anticipated an upper concentration of 100 mg/m³ for particles in the respirable size range.

First (1952) reviewed dust concentrations in industrial atmospheres and noted that some dusts such as wheat flour can reach explosive concentrations at about 20 to 30 g/m³. First concluded that dustiness of this concentration is very unstable because the suspension settles rapidly.

2.4.10 Experimental Investigations

2.4.10.1 Metallic Uranium

Sandia Laboratories (LUNA) conducted explosive tests with metallic uranium using a test device consisting of an ~1-kg hollow cylinder of depleted uranium metal surrounded by shaped explosives. Explosive charge-to-metal mass ratio was the principal parameter of the test series. The values are characterized as low, intermediate, and high. Detonation produced a compact puff of smoke from a combination of explosive combustion products and uranium metal vapors and brilliantly incandescent streamers radiating in all directions for distances up to 200 m. The streamers were particles of uranium heated by self-oxidation. Size and velocity of the metallic fragments were determined from greased plates and an electron microscope that intercepted the radiating particles 1 to 2 m from the event. Seefeldt (1977) developed the information as shown in Table 2.4-11. Particle size distributions fell into two modes: the lower (small particle size), which approximates a log-normal distribution, and the upper (large-particle-size), which does not. The fraction of particles in the lower size range increases as the ratio of explosive to uranium increases. The airborne mass concentrations either were not measured or were not reported. Seefeldt and Mecham (1977) assumed that the initial dispersion cloud had a radius of 300 m, the maximum dispersal distance, and calculated a lower limit average mass concentration of about 10^{-2} mg/m³.

2.4.10.2 Dispersal of Chemical Agents

An extensive experimental program was conducted to study the dispersion of solids (Allan 1970). Resorcinol in two forms was used as a source material; it is a crystalline filament powder ranging from 50 to 600 µm dia, with chips as large as several millimeters. The detonations were performed in a 170-m³ chamber in which the device was centrally suspended. The data obtained were the percentage of the bomblet's initial resorcinol charge remaining airborne versus time.

TABLE 2.4-11. Characteristics of Particles from Explosive Tests That Dispersed Depleted Uranium Metal

	Test		
	Shot 6	Shot 7	Shot 8
Explosive-to-metal ratio(a)	Low	Intermediate	High
Largest particle observed, μm	6250	2500	250
Wt percent of particles in small-particle-size mode	~4.1	~50	~100
Diameter μm , by dividing the small- from the large-particle-size modes	~600	480	250
Data Calculated from Histograms(b)			
Mean diameter μm , based on:			
Number of particles	50	N/A	41
Mass	213	N/A	254
Geometric standard deviation	2.0	N/A	2.2
Data calculated from plot of (dN/dD) versus D(b)			
Mean particle diameter μm , based on:			
Number of particles	41	48	35
Mass	219	159	120
Geometric standard deviation	2.1	1.9	1.9

(a) Actual values were not reported.

(b) These data are for small particle sizes only.

Seefeldt (1977) used the data generated to consider explosive releases in fuel cycle facilities. He examined the effects of three parameters as they relate to size distribution: 1) degree of compaction of resorcinol fill, 2) the presence of confining casing, and 3) test scale. The major findings he made will be discussed below.

1. Compaction: Uncompacted resorcinol yielded the smallest mean particle diameters for all explosive ratios ($ER = \frac{\text{weight of explosive}}{\text{weight of inert}}$). Sizes ranged to 18.5 μm for an ER of 0.2, to 7.2 μm for a more energetic ER of 1.0. Changes in mean particle diameter attributable to compaction could not be considered substantial.

2. Casing: The effect of confinement during an explosive event has been recognized as an important parameter in releases. The addition of casing decreases the effectiveness of dispersion, resulting in larger mean particle sizes. This indicates that an accidental explosion will have the greatest consequences in the absence of confining cases.

3. Scale: Tests were conducted with three different pellet sizes. Differences of mean particle sizes are significant for low ERs (0.2) but essentially disappear for more energetic events (ER = 1.0). For a constant ER, dispersion appears to become less efficient as pellet size increases, as evidenced by larger mean particle sizes.

2.4.11 Pressure Waves

Particles generated by explosions will move with the blast wave, so we must consider how these waves act. Aerosol behavior and transport in enclosures will be considered in a separate report on dynamics and will only be touched upon briefly here.

When a charge is detonated in a building, the initial shock wave is identical with that obtained in the open (White 1946). When this shock wave strikes the walls that surround the charge it is reflected, and the reflected wave bounds back and forth against the walls, floor, and roof until its energy has been completely transformed into heat or until there are no longer confining walls. This is accompanied within the enclosure by a rise in pressure, which can be calculated. If the walls of the room are perfectly rigid and do not conduct heat, without windows or other vents, and if all the available energy of the explosive is realized in the initial detonation and subsequent afterburning, the pressure rise Δp in psi is:

$$\Delta p = \frac{8.8H}{V}$$

where

H = heat of combustion of explosive, kcal (TNT = 3.6 kcal/g)

V = room volume, ft³.

Common to all explosions is the generation of pressure waves (Geiger 1974). Experiences with coal explosions show reflections from the wave produced by the initial pressure rise can result in a reversal in direction of the air movement (Rae 1973). Explosions are very complex phenomena, dependent on the chemical composition of the mixture and geometric conditions.

2.4.12 Liquids

The discussion thus far has been concerned with explosions of solids. This section deals with liquid explosively dispersed, and the information can be applied to sprays also.

The dispersion of liquid in air is a subject of interest to those concerned with chemical warfare agents. Gerber (1976) developed a reusable, vertical, high-explosive projector to simulate the dispersion of chemical agents in a confined space. Eleven liquids, including hot and cold water, were used in the program.

The fraction of liquid below height Z above the projector exhibits a two-parameter Weibull-type distribution as follows:

$$F(Z) = 1 - \exp -\left(\frac{Z}{N}\right)^\beta$$

where

F(Z) = fraction not reaching Z

Z = height, ft

N, β = constants.

For cold water, values N and β were 12.3 and 3.28, respectively.

Seefeldt and Mecham (1978) analyzed reported test data (Dimmick 1958) on the explosive dispersion of an organic liquid. They calculated maximum mass of airborne liquid as a function of the energy of the explosion. The analysis

assumed a log-normal and semilog particle size distribution and calculated the mass in each size fraction. The fraction airborne using both size distribution methods is shown in Table 2.4-12.

Seefeldt (1978) stated that experimental confirmation would be required to choose a model for extrapolation. The log-normal model yields cumulative fractions so low that inhalable quantities can be considered nil. The semilog model yields fractions that are small but nonetheless several orders of magnitude larger than the fractions yielded by the log-normal model. The values are plotted in Figure 2.4-1, comparing the cumulative fraction. It appears that the semilog model yields values that represent upper limits, since a main characteristic of this model is that the number of particles continues to increase without limit as the particle size approaches zero.

TABLE 2.4-12. Comparison of Cumulative Fractions of the Mass of Airborne Material for Log-Normal and Semilog Size Distributions

Upper-Limit Particle Diameter D Defining a Range, μm	Log-Normal Distribution, (a) ($\sigma_g = 2.0$)		Semilog Distribution (a)
	s (b)	p (c)	p (c)
<256	0	5.0×10^{-2}	5.66×10^{-1}
<128	1	1.59×10^{-1}	1.43×10^{-1}
<64	2	2.28×10^{-2}	1.90×10^{-8}
<32	3	1.35×10^{-3}	1.75×10^{-3}
<16	4	3.17×10^{-5}	1.33×10^{-4}
<8	5	2.87×10^{-7}	9.21×10^{-6}
<4	6	9.90×10^{-10}	6.05×10^{-7}
<2	7	1.29×10^{-12}	3.88×10^{-8}
<1	8	6.28×10^{-16}	2.45×10^{-9}
<0.5	9	1.14×10^{-19}	1.542×10^{-10}
<0.25	10	7.77×10^{-24}	9.67×10^{-12}

(a) Modes for both distributions are assumed to be 256 μm .

(b) Number of standard deviations from mode.

(c) Cumulative fraction for size range shown in Column 1.

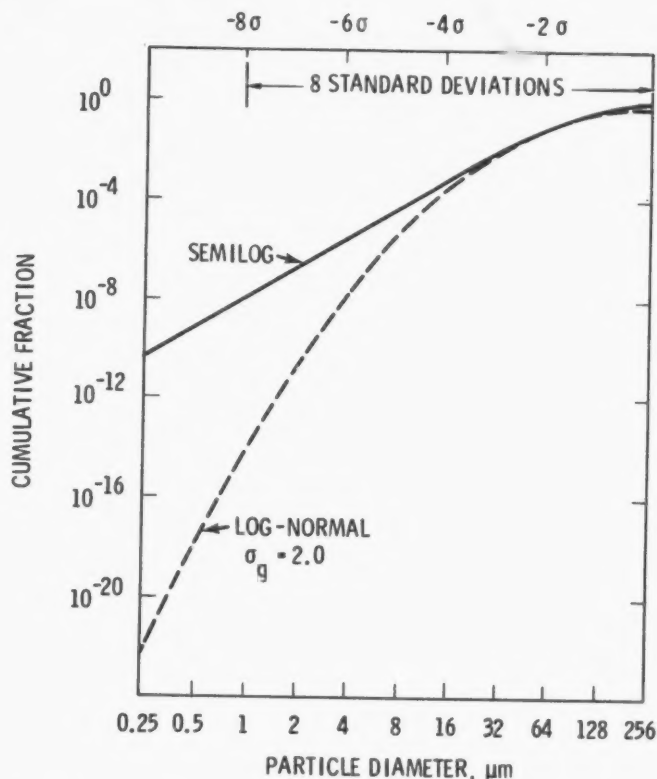


FIGURE 2.4-1. Comparison of Cumulative Fractions of Organic Liquid Airborne for Log-Normal and Semilog Distributions

Seefeldt and Mecham therefore suggested a semilog distribution for liquids, with MMD decreasing as energy-to-mass ratios increase. At 10 cal/g, MMD is estimated at 200 μm . In a semilog-normal distribution, the number of particles as a function of diameter increases as the diameter gets smaller and does not approach zero; while in a log-normal distribution, the frequency of the particle mass (and number) approaches zero as the diameter is reduced.

Seefeldt and Mecham (1979) assembled data on the mechanical energy in cal/g required to subdivide and disperse particulate material during an explosion (included in Figure 2.4-2). The reference energy ratio for the detonation energy of TNT is 1000; for the vaporization of water, 540; and for the adiabatic work to compress air, 67. Coupled with the semilog size distribution suggested above, a mass airborne concentration could be calculated.

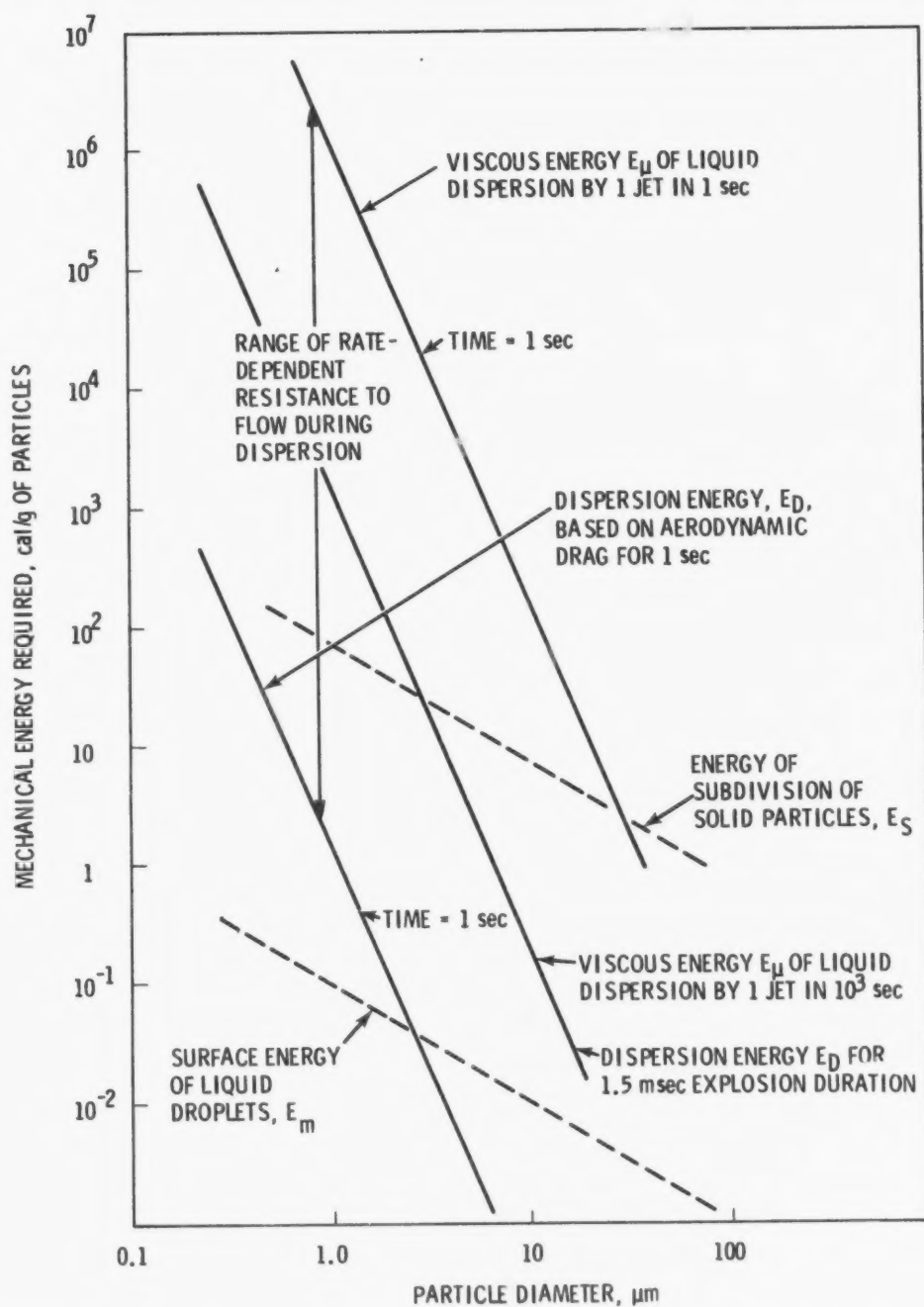


FIGURE 2.4-2. Energy Requirements for Subdivision and Dispersion of Liquids and Solids

Airborne debris could be associated with the particles. Baker (1973) found limited information on fragmentation of bursting liquid propellant vessels: the particles seem to be fairly large and powdered, i.e., into a pound or fraction of a pound, or more small fragments. The fragment velocity increased as a function of blast yield (blast energy, TNT equivalency).

2.4.13 Post-Release Movement of Particles

The respirable particles generated by the explosion would be anticipated to be carried with the airflow. These ejected fine particles would have no mobility of their own, but would follow air currents. Particle movement investigations are not within the scope of this report.

2.4.14 Summary

While explosions have been studied extensively, the information required to estimate particulate material source terms has not been developed. Some estimations of the mass concentration have been made (Selby 1975), but there is a high degree of uncertainty associated with these numbers. Currently, based on this work, 100 mg/m^3 is used as a maximum upper-limit quasi-stable air concentration after an explosive event. Fractional releases of 1×10^{-6} and 1×10^{-7} have been calculated using information generated in two well-documented incidents. This information is insufficient and not reliable enough for accident analysis.

3.0 COMMINUTION

Comminution is described as the pulverizing or dividing of a material into small parts. Mechanical comminution will result from accident-imposed stresses of impact, crushing, grinding, or puncture. Airborne material can be generated by fracture of waste forms such as glass, bitumen, or pellets. The amount and particle size of the airborne particles are required to characterize the release; these would be dependent on the comminution mechanism and material involved in an accident. Some experimental work has investigated impact comminution of waste forms, impacting capsules of fuel pellets, but we could find no work involving contained powders in impact, grinding, or puncture tests.

Primary mechanisms in comminution (Walli 1964) are:

- disintegration shock (shattering)
 smashing by impact
 crushing by pressure
- mechanical shear (abrasive grinding).

Comminution results from the application of energy to some object; in an accident it would be in the form of a drop, impact, compression, or shear.

Various measures can be used to describe the application of energy. For example, the joule (J) is the meter-kilogram-second unit of work or energy, equal to the work done by a force of 1 newton (N) when its point of application moves 1 m in the direction of the force. Other units can be used to describe the energy using its tendency to turn into heat. There is an exact relationship for the mechanical equivalent of heat:

$$1 \text{ calorie} = 4.1833 \text{ international joule}$$

Other energy descriptors which can be used to describe the input energy in the comminution process include Btu, calorie, electron volt, erg, foot pound, etc. Some of these terms are used in the reports cited, but no attempt to convert to one uniform measure has been made, since there is apparently no commonly accepted unit.

Both solids and powders can be subject to comminution resulting from an accident in a fuel cycle plant. For example, stresses on powders in press-metal cans could rupture the cans, exposing the powder to comminution and airborne release. This chapter will cover both solids and powders responding to comminution stresses. Data from both current industrial processes and experimental work will be used.

3.1 INDUSTRIAL PROCESSES

Industrial crushing and grinding operations (Herdan 1960) were investigated to give insight into the process of comminution. In crushing and grinding, particles are subject to a mechanical force and they break into more fine-grained particles. Grinding is based on fracture; the force is abrading so that the grain is gradually worn to pieces. Crushing, on the other hand, results from pressure alone.

Mill grinding of a product produces grains of unequal size. Log-normal particle size distributions should be produced by grinding, and it has been found empirically that for many materials this is the case. This cannot be completely generalized, however, because of differences in materials and grinding equipment. Fineness of various materials from different equipment processes is illustrated in Table 3.1-1. Roughly speaking, machines which work by crushing action only, like jaw crushers, will give a smaller weight percent of fine particle sizes than those combining crushing and abrasion. In ball mills, the smaller the balls, the finer the material. Most of the material was fairly coarse; however, all the particles in the smallest size group, 0.02 mm, were $<20\text{ }\mu\text{m}$. Herdan's work did not attempt to look at the respirable range, but indications are that it was small.

Grinding of a solid increases its particle surface area. The amount of new surface created per unit weight (ft/lb) per unit time serves as a measure of grindability. The surface area produced by crushing and grinding is directly proportional to the energy absorbed or consumed.

These observations indicate the necessity of describing the material and force to explain accident-associated comminution releases.

TABLE 3.1-1. Fineness of Materials Produced by Different Grinding Equipment

Material	Maximum Particle Diameter, mm			
	Jaw Crusher	Roller Mill	Disintegrator	Ball Mill Steel Balls, Dry Grinding
Glass	18	15	1.0	0.45
Feldspar	14	9	0.5	0.42
Flint	9	4	0.5	0.09
Brick	5	4	0.4	0.04
Glance-iron	4	1	0.2	0.02

It might be expected that the rate of breakage during comminution will increase with time, as has been indicated by studies of grinding operations (Berube 1979). This was observed in experiments on breakage of quartzite. Insufficient particles $\leq 10 \mu\text{m}$ were generated, however, to be detectable in the particle size distribution.

Abrasive blast cleaning (Danielson 1973) produces airborne dust concentrations. The abrasives are propelled by compressed air, high-pressure water, or centrifugal force. Propelling pressures for compressed air were not noted; water and sand are propelled through a nozzle that develops pressures of 1000 to 2000 psi. Typical particle size analysis of emissions produced using shot or sand is shown in Table 3.1-2. About 15 percent and 22 percent of the particles were $<10 \mu\text{m}$ for steel shot and sand blasting, respectively. Since the airborne portion included the cleaner (already dispersed) and the abraded material, these could be upper limit particle size values for abrasive comminution of a solid surface area.

3.2 SOLID WASTE INVESTIGATIONS

Concretes and glasses considered for incorporation of radioactive waste sludge were subjected to impact tests to determine the relationship between the energy of the impact and the resulting increase in surface area of the sample (Wallace 1976). Cylindrical specimens were impacted with a 2-kg weight dropped from known heights, at a maximum energy of 1.6 kg-m (15.7 J). A particle size analysis was performed on the crushed material. Successive

TABLE 3.1-2. Particle Sizes Produced by Abrasive Cleaning

Steel Shot		Sand	
Particle Diameter, μm	Cumulative Weight-Percent	Particle Diameter, μm	Cumulative Weight-Percent
1.7	1.4	1.6	0.4
2.8	4.7	2.7	1.8
4.5	7.8	4.4	5.3
6.7	10.3	6.6	11.1
8.4	12.2	8.3	15.6
9.5	13.4	9.3	18.2
10.6	14.8	10.4	21.6
11.7	16.0	11.5	24.6
15.1	19.3	14.8	33.2
20.1	23.0	19.8	41.3
26.8	28.6	26.4	50.1
33.5	35.7	33.0	58.7
41.9	44.3	41.2	65.6
50.3	52.1	49.5	72.1
67.0	62.7	65.9	82.0
100.6	73.1	98.9	96.5
150.3	80.5	147.8	100.0

crushing of the same sample was continued until different energy totals were reached. The resulting particle sizes generated are listed in Tables 3.2-1 and 3.2-2, and plotted in Figures 3.2-1, 3.2-2, 3.2-3 and 3.2-4.

The crushing of glass produced fewer fine particles that are likely to become airborne than did the specimens of concrete. In crushing with 8 kg-m of energy per 10 g of sample, none of the glass particles were less than 8 μm .

Under only slightly more severe conditions (9.6 kg-m per 10 g), 0.016 percent to 0.032 percent of the neat concrete samples, and 0.2 percent to 0.4 percent of the concrete samples containing sludge were converted to particles <4 μm . These are very small fractions that are likely to become airborne.

TABLE 3.2-1. Particle Size Distribution of Glass Crushed by Impact

Particle Size (x), μm	Cumulative Percent with Particle Size Less than x			
	2 kg-m	4 kg-m	6 kg-m	8 kg-m
2000	26.4	51.2	70.3	81.9
1000	14.2	33.4	47.8	57.4
500	7.4	19.5	31.1	38.6
250	3.7	10.8	18.9	23.4
125	1.7	5.5	9.9	12.1
62	--	--	--	6.2
33	--	--	--	1.24
16	--	--	--	0.11
8	--	--	--	0

Smith (1975) performed impact tests on vitreous wastes, where encapsulated waste forms were smashed at velocities up to 177 fps. The waste canisters were breached in small cracks, and little, if any, glass escaped. The maximum fraction $<10 \mu\text{m}$ was 10^{-4} .

Bunnell (1977) examined various factors relevant to the thermal and mechanical shock resistance of waste-containing glasses. Soda-lime-silica plate glass specimens 0.440 in. dia and 0.5 in. high were placed in a die and impacted by a falling weight. After crushing, the glass pieces were sized by sieve analysis. Figure 3.2-5 shows the size distribution as a function of impact energy. The principal effect of higher energies is to increase the fraction of fines, and the curves represent the average of several impacts.

Particle size distribution is a rather strong function of the number of impacts; Figure 3.2-6 shows particle size produced by the same total amount of energy (158.4 ft-lb) applied as a single impact or up to eight impacts. With the multiple impacts, the particles were stirred after each impact. A large difference exists between single and multiple impacts, particularly in comparing one, two, and three impacts. At greater numbers of impacts, the bed of particles probably distributes the energy so that point loading effects decrease. The $<10 \mu\text{m}$ fraction was about 4 percent for eight impacts.

TABLE 3.2-2. Particle Size Distribution of Concretes Crushed by Impact

Particle Size (X), μm	Cumulative % with Particle Size Less Than X											
	High Alumina (Neat)		Type IP (Neat)		Type III (Neat)		High Alumina + 40% Sludge		Type IP + 40% Sludge		Type III + 40% Sludge	
	4.8 (a)	9.6 (a)	4.8 (a)	9.6 (a)	4.8 (a)	9.6 (a)	4.8 (a)	9.6 (a)	4.8 (a)	9.6 (a)	4.8 (a)	9.6 (a)
2000	20.9	49.6	38.1	58.0	34.1	63.6	49.3	70.6	54.1	76.5	50.6	63.4
1000	9.3	27.2	17.4	32.5	14.4	36.5	29.8	50.8	30.8	53.2	30.5	43.1
500	4.9	16.1	8.7	17.6	6.4	19.5	19.3	36.0	16.7	33.7	20.2	29.5
250	2.8	9.8	4.3	9.7	3.4	10.5	12.9	25.0	9.4	19.8	13.1	20.3
125	1.4	4.2	2.3	5.5	2.0	5.5	4.5	16.1	5.0	10.8	8.3	12.5
62	--	2.1	--	2.8	--	--	--	10.3	--	6.2	--	8.8
33	--	0.71	--	1.27	--	--	--	4.7	--	2.7	--	5.5
16	--	0.21	--	0.48	--	--	--	2.2	--	1.03	--	2.4
8	--	0.057	--	0.15	--	--	--	0.90	--	0.38	--	0.63
4	--	0.034	--	0.072	--	--	--	0.40	--	0.21	--	0.20
2	--	0.016	--	0.032	--	--	--	0.16	--	0.11	--	0.084
1	--	0.0	--	0.0	--	--	--	0.03	--	0.03	--	0.014
0.6	--	0.0	--	0.0	--	--	--	0.0	--	0.0	--	0.0

(a) kg-m

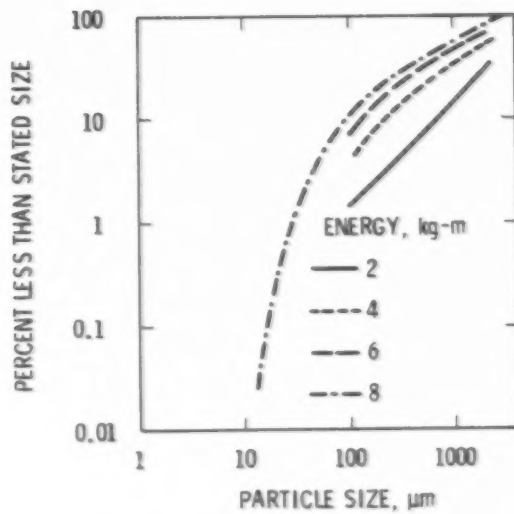


FIGURE 3.2-1. Particle Size Distribution of Glass Crushed by Impact

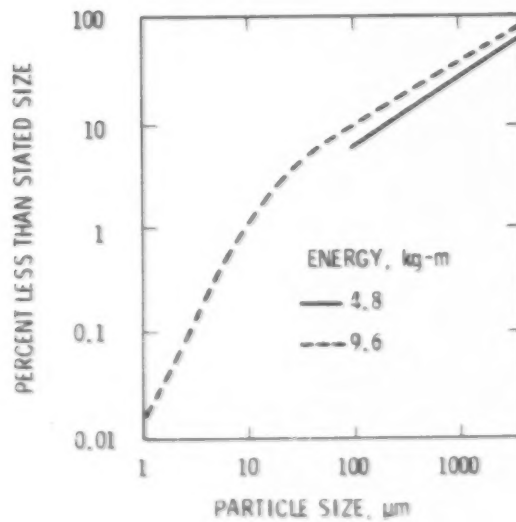


FIGURE 3.2-2. Particle Size Distribution of Type III Concrete Crushed by Impact

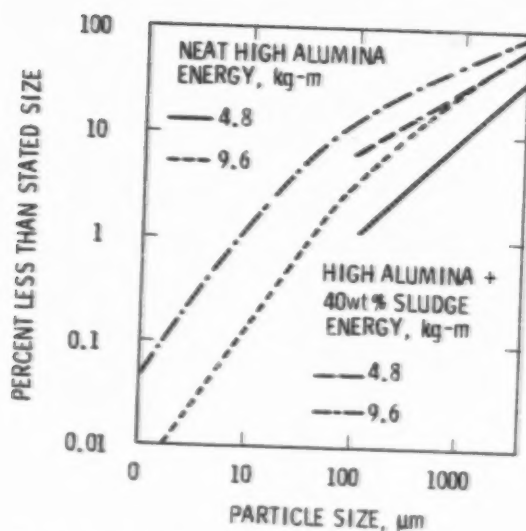


FIGURE 3.2-3. Particle Size Distribution of High Alumina Concrete Crushed by Impact

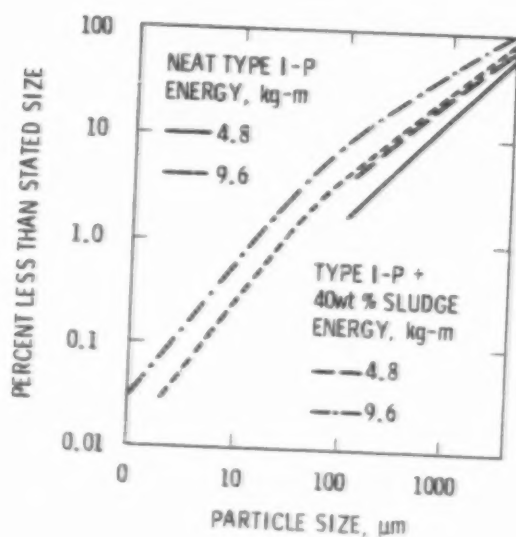


FIGURE 3.2-4. Particle Size Distribution of Type I-P Concrete Crushed by Impact

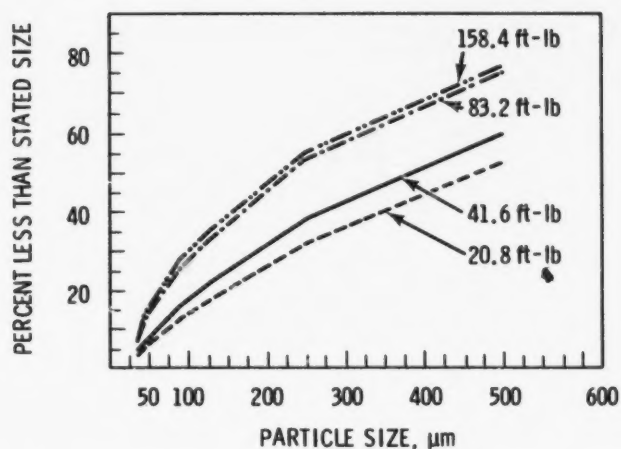


FIGURE 3.2-5. Averaged Size Distribution Curves for Soda-Lime-Silica Glass Impacted at Four Different Energies

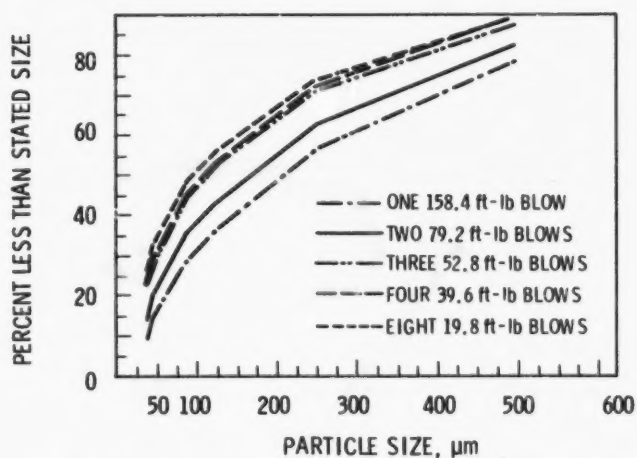


FIGURE 3.2-6. Size Distribution for Soda-Lime-Silica Glass Impacted by the Same Total Amount of Energy, 158.4 ft-lb, Applied as Single or Multiple Blows

Mecham (1979) presented a method for analyzing particle size distributions obtained in impact testing of brittle-waste materials. He compared his results with impact test data generated by other investigators, and concluded that particles produced in impact fracture of brittle materials have a linear correlation in a log-normal particle size distribution with a nearly constant brittle-fracture surface-energy factor. The method allows estimation of total surface area of particles from impact fracture. Reliable extrapolation of particle size distributions to any log-normal range seems achievable.

Mecham developed an equation relating the particle surface area to solid volume (S_F/V_0) as follows:

$$\frac{S_F}{V_0} = \frac{\alpha(\sigma_g)^{0.5 \ln \sigma_g}}{D_g}$$

where

S_F = surface area

V_0 = solid volume

α = dimensionless shape factor with a minimum value of 6 for spheres and cubes, two or three times higher for other shapes

D_g = geometric mean diameter

σ_g = geometric standard deviation.

The total particulate surface area S_F is related to the original specimen surface area S_0 and to the surface area formed in a fracture as:

$$\Delta S_F = S_F - S_0$$

Fracture energy W_F absorbed in the brittle material is proportional to S_F over a range of energy input for a given specimen and mode of impact. The proportionality constant is termed the fracture surface energy γ_F :

$$W_F = \gamma_F \Delta S_F$$

Fracture energy W_F is generally smaller than the gross impact-energy input W_I because of deformation in the equipment. Generally it is about ≤ 50 percent of W_I . Variations of γ_F can be expected for different materials, sizes and shapes, and for variations of input energy concentrations in specimen volumes (V_0). Experimental work indicates this equation may be used to estimate ΔS_F for an impact energy W_F , since γ_F is reasonably constant at about 100 J/m^2 . Work is continuing to confirm this methodology.

All of the experimental work with waste forms indicates only a small amount of the material will be in the respirable fraction of $<10 \text{ }\mu\text{m}$.

3.2.1 Breakup of Fuel Forms

Capsules containing microspheres of the fuel plutonium dioxide ($^{238}\text{PuO}_2$) were impacted into concrete, granite, and sand targets at velocities from 102 to 456 fps (Bonnell 1967, 1969). The fines produced were analyzed. The damage to the microspheres and percentage of fines produced from an end-on impact were determined. The percentage of fines ($\leq 44 \text{ }\mu\text{m}$ dia) increased with velocity to between 40 percent to 55 percent. The maximum respirable percentage $<10 \text{ }\mu\text{m}$ was about 13 percent for the 456 fps impact on concrete. Other values for lesser impact ranged downward from about 6 percent to not detectable. Before impact, the fraction $\leq 20 \text{ }\mu\text{m}$ was not detectable and the MMD was about $100 \text{ }\mu\text{m}$. This work indicates that impact can increase the fine particle fraction of contained materials considerably.

3.2.2 Shock Compression

When a material is compressed by a shock wave, the energy can be transformed into kinetic energy of flying fragments (Dremin 1971). Pulverizing action of shock waves has been observed. Friable materials can be comminuted to particles of smaller size than obtained by other methods. For example, silicon carbide, when ball-milled for a period of time, is reduced to 100 to $350 \text{ }\mu\text{m}$ particles. If this same material is subjected to shock compression, the particle size does not exceed 0.5 to $0.6 \text{ }\mu\text{m}$. Dremin was interested in shock compression 500 kbar or about $7.3 \times 10^6 \text{ psi}$. Thus, these observations are of general interest, but appear to be much more extreme conditions than are likely in fuel cycle facility accidents.

3.2.3 Comminution of Coal

Coal size distributions of coarse fragments and fine particles resulted from different modes of energy application--drop, impact crushing, wedge impact, compression and shear tests (Kurth 1975). Airborne dust and respirable airborne dust distributions were obtained from impact crushing and wedge impact tests. As with other comminution investigations, the fraction in the small size fraction increases with increasing energy.

Fourteen impact-crushing pendulum tests produced valid data. The total number of dust particles in various size ranges was collected in the first minute after breakage. Tests were conducted at 95, 164, and 293 ft-lb energy levels. The most respirable particles were produced with the greatest application of energy.

The investigation of impact tests determined that the respirable dust counts can be predicted from the nonairborne^(a) particles in the respirable range (for Kurth's work, $\leq 5 \mu\text{m}$). Letting N denote the respirable dust count, and n denote the nonairborne particle count, a power law of the form $N = an^b$ was used to fit the data. Power law coefficients are shown in Table 3.2-3. These coefficients can be used in a linearized power law equation to predict the respirable dust count for the first minute after impact either from impact energy or specific crushing energy (ft-lb/ft^3).

TABLE 3.2-3. Power Law Coefficients for Relating Airborne Respirable Dust Counts to Nonairborne Dust Counts

Impact Energy, ft-lb	a	b
95	1,747	0.1327
164	3,238	0.1031
293	4,193	0.1026

(a) Coal particles in the size range that do not become airborne from an event.

Conclusions from the various tests were:

- Impact pendulum crushing--The Weibull distribution showed the best fit of six tested distributions. The percent of fine particles increased with level of energy applied. The number of particles of respirable dust increased with input energy but the respirable dust distributions remain approximately the same for any level of energy input.
- Impact pendulum wedge--The Weibull distribution showed the best fit to nonairborne dust in the respirable size range, and was the same for five different input energies.
- Compression and shear tests--The Weibull distribution showed the best fit.

In cutting coal, the dust/g of broken coal produced from single cuts decreases as cut depth increases (Strebig 1975). For a 0.25 in.-deep cut the maximum particle size was 1.0 $\mu\text{g/g}$ for Illinois coal or 9.4 $\mu\text{g/g}$ for Pittsburgh coal. For 2 in.-deep cuts about 0.07 μg of respirable dust/g of coal was generated. This implies that a puncture accident would generate very little airborne dust. Once again, the material involved in an event will be an important consideration.

In other work with coal, Hamilton (1958) found the breakage products of a variety of coals could be expressed by:

$$\frac{dF_N}{dD} = \alpha D^{-\beta}$$

where

dF_N = number of particles in the size range from D to $D + dD$

α = constant, not specified

D = particle size

β = constant.

For coal and rock the values of β are 2.25 and 3, respectively. This relationship was found to be reasonably valid in the size range 1 to 100 μm for dust-producing operations such as rotary and percussive drilling, coal cutting and shatter.

In studies of explosions in stone blocks, Grimshaw (1958) concluded that the size distributions generated followed the Hamilton model.

3.2.4 Summary

This review of comminution events indicates that some relationships have been experimentally discovered between energy input and particle generation from mechanical comminution, and in the case of coal, relationships between energy and the airborne fraction. However, the relationship between the materials used in these studies and those potentially involved in fuel cycle accidents has not been established. It was established that releases between material types--for example, two types of coal--are different. Therefore, the energy/particle size relationships discussed can only give a rough estimate of airborne releases.

The literature reviewed indicated that releases of respirable particles from comminution events would be low. One area of interest where no information was found was measurements of contained or uncontained powders subjected to impact or crushing tests.

4.0 BEHAVIOR OF LIQUID PARTICLES

Some aspects of liquid particles have been alluded to in earlier sections. However, unique phenomena such as evaporation are associated with airborne liquids and require special consideration.

Liquid aerosols are typically comprised of spherically-shaped droplets that can bounce, slide, and spread after impact. Droplets can grow and shrink by diffusion of vapor to and from the surfaces (Dennis 1976).

Nuclear fuel cycle accidents can generate liquid aerosols by leaks, spills, sprays, impacts, flashing sprays, and chemical reactions. In assessing an accident, we are interested in estimating how much liquid particulate becomes airborne and the particle size generated. Empirical studies generally do not present source-term mass information in a straightforward manner, such as weight percent of source airborne. This is probably due to a lack of interest in the particular information because many studies of liquids, particularly sprays, have been made. For example, one study developed a distribution between the mass in a spray and the mass leaving a spray. It appears that a great deal of this information could be extrapolated for use in estimating accidental releases. However, the number of variables associated with any accidental release made it impractical to identify a single method. Different formulations may be appropriate for different situations.

Two bodies of information that would seem useful for developing source terms are:

- Sprays and atomization studies that have empirically developed equations for estimating the particle size and mass distribution of a release. Some pressurized liquid releases will be sprays, and this information would be useful.
- Traditional fluid flow relationships such as the Darcy formula for pipe flow that could be used to estimate the volume passing from a breach.

In the discussion of liquid particles, particularly jets and sprays, the term Weber number is used. It describes the disintegration of liquid particles (sprays) by surface tension forces and aerodynamic stresses (Brown 1961). One form of the Weber number is

$$N_{We} = \frac{\rho_g V^2 d}{2 g_c \sigma}$$

where

We = Weber number

ρ_g = density of gas phase

V = velocity of spray relative to gases

d = diameter of liquid jet

g_c = conversion factor (poundals per pound force)

σ = surface tension.

The Weber number is the ratio of the impact stress of the gas phase on the interface to the normal stress caused by the interfacial tension acting on any cross section. For low-viscosity fluids the type of disintegration depends on the Weber number. When $We < 0.2$, only the pinching-off action of interfacial tension applies; from $0.2 < We < 8$ the action is a serious distortion which whips the jet into segments, and for $We > 8$ the action is more violent, with ligaments of fluid separating from the jet and atomization occurring. At even higher Weber numbers the masses and drops of liquid formed originally from the main jet will themselves be broken up further; i.e., secondary atomization will occur.

Sprays can mitigate the consequences of an accident; for example, dust can be suppressed by water scavenging of air. This is acknowledged but not detailed since it is not within the scope of this study.

General areas considered are:

1. leaks and spills
2. entrainment of droplets
3. sprays

4. flashing sprays
5. evaporation
6. liquid pool releases
7. sparging.

Condensation aerosols are considered in a separate section.

Although a considerable body of information has been developed associated with loss-of-coolant reactor accident (LOCA) releases, this type of event has not been considered here since it is an accident that would not occur in other fuel cycle facilities.

4.1 LEAKS AND SPILLS

No references were found that reported measurements of the airborne release from leaks or spills. Therefore, estimations of airborne fog and droplet concentrations have been used to estimate airborne releases from a spill.

At superficial air velocities of less than 0.046 m/sec (0.15 fps), aerosols formed by vigorous mixing of a solution in air are metastable and have a concentration of $\sim 10 \text{ mg/m}^3$ (ORNL 1970). This metastable concentration is somewhat equivalent to a fog, which is about 10 mg/m^3 with a particle size of approximately $10 \text{ }\mu\text{m}$. Garner (1954) reported that the weight distribution of particles smaller than 10 to $20 \text{ }\mu\text{m}$ will be fairly constant even if there is gross entrainment of larger droplets.

This knowledge may be used to estimate the concentration of particles smaller than a given size, even in an airstream which is very concentrated with liquid droplets. Liquids in air can have higher concentrations; for example, a 1 in./hr rain with a mass mean particle size of $3000 \text{ }\mu\text{m}$ has a concentration of 1 g/m^3 . In natural clouds the liquid water content is $\sim 1 \text{ g/m}^3$ (Fuchs 1964). The 10 mg/m^3 value can be used in estimating inhalation hazard associated with an operation since it reflects the concentration of particles $<10 \text{ }\mu\text{m}$ dia.

These values are for water droplets; in a nuclear fuel cycle facility, other liquids such as acids or solvents could produce aerosols. Airborne acid concentrations might range from 1 to 10 mg/m³, and a volatile solvent such as acetone could reach much higher levels, such as 1 x 10⁵ mg/m³ (Bond 1972). These are vapor concentrations that could produce aerosols on cooling.

The flow of liquids through nozzles and orifices can be determined from the following established and accepted formula (Crane 1977):

$$q = 3.48 \times 10^{-6} d_1^2 C \sqrt{h_L} = 3.51 \times 10^{-4} d_1^2 C \sqrt{\frac{\Delta P}{\rho}}$$

where

q = flow rate, m³/sec

d_1 = nozzle or orifice diameter, mm

C = discharge coefficient^(a)

h_L = loss of static pressure

ρ = density, kg/m³

ΔP = pressure differential between upstream and discharge points, bar.

These relationships could be used to estimate total liquid discharge from a breach if the size of the opening can be estimated for a spill.

4.2 ENTRAINMENT OF DROPLETS

Droplets are formed by:

- Splashing. This occurs in any liquid/vapor system and is dependent on vapor volume and surface area. Splashing increases in vapor passage per unit area of interface; if agitation is more vigorous, there is more entrainment.

(a) A function of nozzle geometry and flow Reynolds number R_C (e.g., a sharp-edged orifice opening in a large space has $C = 0.7$ and $R_C = 20$, which decreases to $C = 0.6$ for $R_C \geq 1000$).

- Bubble bursting. The bubble rises to a liquid surface and forms a dome-shaped lamina that ruptures. It has been reported that a cloud of droplets 60 μm dia is formed (Knelman 1954).
- Foaming. As a bubble rises through a solution, absorption at its surface may produce a concentration gradient between bubble surface and the bulk of the liquid; electrical potential differences will therefore be established and will inhibit coalescences of rising bubbles. Clouds of small bubbles reaching the liquid surface produce a foam. Droplets projected from a foam layer come from the rupture of bubbles. Entrainment rates under such conditions should be fairly constant.

Ninety-five percent of the droplets entrained in the vapor space of evaporators were $<20 \mu\text{m}$ dia, but because of their low mass they formed only a very small fraction of the weight of the entrained liquid (Garner 1954).

Droplets will break up in a gas stream due to aerodynamic forces if the Weber number is higher than the critical value. This phenomenon was studied by Krzeczowski (1979). He found that the mechanism of droplet deformation and disintegration as well as break-up duration depends on the Weber number, Laplace number and the ratio of liquid viscosity to gas viscosity. The influence of the Weber number is the strongest. Viscosity only weakly influences the studied conditions; e.g., while viscosity increases by about 10^3 , breakup duration of liquid droplets due to an airstream increases only twice.

4.3 SPRAYS

Spraying and atomizing are terms describing the process in which liquids are transformed into droplets dispersed in air or other gases. Spraying refers to the formation of coarse droplets, atomizing to the production of fine droplets. Spraying and atomizing, which have been studied extensively (Orr 1966), can involve several physical processes.

4.3.1 Physical Processes

4.3.1.1 Jet Disintegration

A stream of an inviscid liquid discharging from a round opening into a gas at relatively low velocity will most likely have a length 4.5 times the diameter of the jet. The droplet diameter can be geometrically calculated as slightly less than twice the diameter of the opening producing the jet. Some factors in jet disintegration are:

- Liquid Viscosity. Low-viscosity liquids with high interfacial tensions injected at low velocity give droplet sizes averaging around three to four times the initial jet diameter. As liquid viscosity increases the most probable jet-disintegration length increases, reaching as much as 40 times the initial jet diameter for castor oil, contrasted to 4.5 times the diameter for water. Increasing jet velocity results in shorter breakup lengths.
- Aerodynamic Forces and Turbulences. As soon as any disturbance appears in a stream wave, motion is intensified, and the stream can be pinched off into droplets.
- Turbulent Eddies in Liquid. These eddies are increasingly important as injection velocity of the liquid increases. A fuel oil stream at 20 psi, for example, will form droplets, in accordance with theory. If the injection pressure is increased to 100 psi the stream will tear into a number of small irregular ligaments which break up into droplets. At 200 psi the stream is likely to shift into an undulating column from which ligaments of liquids are drawn off.
- Satellite Droplets. Theory predicts that satellite droplets will be produced when a jet breaks into droplets. Satellite droplets are often an order of magnitude smaller. Satellites can be 1 to 2 μm dia from spray droplet beads to as large as 2 mm dia.

4.3.1.2 Secondary Breakup

Droplets are broken up immediately if they retain velocity or encounter a fast-moving gas stream. In the case of a comparatively large drop exposed to

a steady stream of air, the drop becomes increasingly flattened, and at a critical velocity is blown into a hollow bag having a roughly circular rim. The bag burst produces a shower of very small droplets; the rim, containing a major portion of the initial drop's mass, divides later into a number of larger droplets.

Drops subjected to very high velocity airblasts maintain a convex surface that is presented to the oncoming air. From the edge, liquid is drawn out into thin sheets and fine ligaments or filaments that disintegrate into droplets. The process is rapid, but not instantaneous; the drop accelerates while bits of liquid are continuously being torn from it. As the relative velocity between the air and drop diminishes, the thickness of the layer being stripped from the drop decreases. A wide range of droplet sizes is thus accounted for because droplets formed from the thin films of the early stages of breakup must be considerably smaller than those formed from the thicker film.

4.3.1.3 Impaction

When a liquid droplet collides with a solid surface at a relative velocity of up to 1000 ft/sec, the droplet first spreads as a flat disk over the solid. The perimeter of the disk subsequently breaks into radial filaments and ultimately into a mist of fine droplets. In the case of a water droplet initially of about 2 mm dia, the mist droplets range from 4 to 25 μm dia.

4.3.2 Size Distribution

It can be seen from the above discussion that sprays can result in a range of particle sizes, depending on the conditions. Marshall (1954) discusses many of the theoretical aspects of spray formation where aerodynamic forces and surface tensions are key forces in disintegration. The size distribution of drops formed by sprays could be relatively coarse (Brown and York 1961). In measurements of the spray from a specific nozzle (Sanford 1975), the maximum drop size was 3832 μm ; 0.2 percent were <25 μm and about 0.1 percent were <20 μm .

Mechanical atomization of liquids in atomizers, nozzles, etc., gives, as a rule, very polydisperse mists (Davies 1966). Atomization proceeds as follows: under the action of hydraulic pressure, a centrifugal or an aerodynamic

force, the liquid is drawn into narrow ligaments or films which subsequently disintegrate into droplets under the action of surface tension. The thinner the liquid ligament, the smaller the droplets formed, but the degree of dispersal always remains large. When liquid flows slowly out of a capillary tip, drops of a constant size are formed, accompanied by very small droplet-satellites.

These examples show the range of particle sizes that can be produced by sprays. Calculations have been developed to estimate the drop diameter. Because these methods are well established, we have not given all the correlations. Perry's Chemical Engineer's Handbook (1973 or a more recent edition) is suggested as a fundamental reference for further calculations for specific sprays.

Well-defined distributions are applicable to various systems, depending on the mechanism of disintegration involved (Fraser 1956). Some of the most frequently used for drops are:

4.3.2.1 The Rosin-Rammler Volume Distribution

This equation is applicable to sprays from swirl nozzles. Its algebraic form is:

$$V_d = 100 \exp\left(\frac{-d}{\bar{d}}\right)^q$$

where

d = the diameter of the drop

$\bar{V}_d$ = the volume percent oversize

$\bar{d}$ = the Rosin-Rammler mean

q = the dispersion coefficient

If this equation is applicable to the actual drop size distribution from a spray, a graph of $\log (100/\bar{V})$ against d on log-log paper should give a straight line. The slope of this line, q , is the dispersion coefficient. The value of

q will lie between 2 and 4, the higher values indicating a more uniform distribution. A portion of the spray volume, 36.8 percent, lies above the Rosin-Rammler mean, $\bar{d}$. This mean can therefore be read from the size distribution curve. Other parameters can be calculated from those used in the equation (Mugele 1951).

4.3.2.2 The Nukiyama-Tanasawa Number Distribution

This equation has been derived for spray distributions from twin fluid atomizers. It has also been used for determining drop distributions obtained from the shatter of drops by air blast. Its algebraic form is:

$$\Delta N = a d^2 \exp(-bd^q)$$

where

ΔN = the number of drops in size group, $d \pm \Delta d/2$

a, b = constants.

If this equation is applicable to a given drop size distribution, a graph of $\log (\Delta N/d^2)$ against d^q gives a straight line whose slope is a measure of d_s (Sauter mean diameter). The dispersion coefficient q is a constant for a given nozzle over a wide range of conditions and is determined by trial and error. It varies from 1/6 to 2, high values indicating a narrow distribution. Other parameters can be calculated from the slope of such a graph or directly from the equation.

4.3.2.3 The Log Number Distribution

An equation to represent the size distribution of drops obtained from a twin fluid atomizer is:

$$\Delta N/\Delta d = a N \exp(-ad)$$

where a is a form of a mean diameter of the spray. If this equation is applicable, a graph of $\Delta N/(N \cdot \Delta d)$ against d on log paper gives a straight line, the intercept and slope of which are measures of the mean diameter. No simple measure of the size dispersion can be obtained.

4.3.2.4 The Log-Normal Number Distribution

The log-normal volume distribution has been used for drops obtained from the spray atomizer. This is one of the most frequent distribution laws occurring in nature. Its algebraic form is:

$$\Delta N = \frac{N}{\sqrt{2\pi} \log s_g} \exp \left[\frac{(\log d - \log \bar{d}_g)^2}{2 \log^2 \sigma_g} \right]$$

where

N = the total number of drops

$\bar{d}_g$ = the geometric drop size mean

σ_g = the geometric standard deviation

and

$$\log \sigma_g = \sqrt{\frac{\sum_{i=1}^n \log^2 (d / \bar{d})_g}{n - 1}}$$

where n = the number of drop size groups. If this equation is applicable to the given drop size distribution, a graph of d against N_d (the number percent oversize) on log-probability paper gives a straight line where the geometric mean $\bar{d}_g$ corresponds to $N_d = 50$ percent and σ_g is the size ratio corresponding to N_d at 50 percent and N_d at 15.93 percent. The advantage of this distribution is that the dispersion factor is in a form which can be handled according to well-established laws of statistics.

4.3.2.5 The Root Normal Volume Distribution

This equation has been applied to drop sizes from swirl sprays. It is similar to the log-normal number distribution above, except $\log d$ is replaced by $\sqrt{d}$, the number parameters N_d are changed in volume parameters $\bar{V}_d$, and s_g and $\bar{d}_g$ are interpreted as the basis of $\sqrt{d}$.

4.3.3 Breakup

Hinze (1955) found that breakup by hydrodynamic forces can be nearly characterized by the Weber number. When one fluid is squirted into another, the fluid is at first present in bulk. The mechanism that is common to disintegration of the bulk fluid is the penetration of ligaments of one fluid into another. These in turn break up into globules which further split up into smaller parts. The size of the droplets seem to depend on many uncontrollable conditions, and distribution cannot be calculated theoretically. Predictions can be made from empirical correlation; however, breakup of drops will have different mechanisms according to flow, viscous shear flow, air flow, turbulent flow, and can be calculated.

4.3.3.1 Viscous Flow

The deformation of globules in viscous flow is determined by the Weber number:

$$We = \frac{\mu_c S D}{\sigma}$$

where

We = Weber number

μ_c = absolute viscosity

c = continuous phase

S = maximum velocity

D = globule diameter, cm

σ = interfacial tension, dyne/cm.

4.3.3.2 Airstream Breakup

The Weber number for airstream breakup is:

$$We = \frac{\rho_c U_o^2 D}{\sigma}$$

where

We = Weber number

ρ_c = density, g/cm²

c = continuous phase

U_0 = maximum relative velocity with respect to drop, cm/sec

D = drop diameter, cm

σ = interfacial tension, dyne/cm.

4.3.3.3 Turbulent Flow

Breakup of drops in turbulent flow has the following relationship:

$$We = \frac{\rho_c \bar{V}^2 D_{\max}}{\sigma}$$

where

We = Weber number

ρ_c = density, g/cm²

$\bar{V}^2$ = average value of square of difference in turbulence velocity,
cm²/sec²

$D_{\max}$ = largest drop size, cm

σ = interfacial tension, dyne/cm.

The maximum drop size shows this relationship:

$$D_{\max} \left(\frac{\rho_c}{\sigma} \right)^{3/5} \epsilon^{2/5} = \text{constant}$$

where

$D_{\max}$ = largest drop size, cm

ρ_c = density, g/cm²

σ = interfacial tension, dyne/cm

ϵ = energy input per unit mass and time, or dissipation per unit
mass, cm²/sec³.

Ingebo (1968) was able to correlate drop size data for a circular jet entering a gas phase with the equation:

$$\frac{D_o}{D_m} = 0.64 \left(\frac{D_o}{D_t} \right)^{0.33} Bo^{0.33} + Re_L^{-0.1} Re_g^{0.5} Bo^{-0.77} \\ \times \left[0.044 + We_L We_g^{-0.2} Bo^{-0.07} (1.25 \times 10^{-6} + 0.33 \times 10^{-6} Ac^{0.5} Bo^{-0.67}) \right]$$

where

D_o = orifice diameter

D_m = diameter of largest drop

Re_L = Reynolds number based on liquid properties,

$$Re_L = \rho_L D_o V / \mu_L$$

Re_g = Reynolds number based on gas properties

Ac = aerodynamic acceleration to surface-tension force ratio based on orifice diameter, $Ac = \rho_g D_o^2 a / \sigma$, where a = absolute gas stream acceleration, cm/sec^2

We_L = Weber number based on liquid properties,

$$We_L = \rho_L D_o (V)^2 / \sigma$$

We_g = Weber number based on gas properties

Bo = Bond number, $\rho_L D_o^2 g / \sigma$

ρ = density of fluid

V = relative velocity

μ = fluid viscosity

σ = surface tension

O, L, g = orifice, liquid, and gas, respectively

t = tip.

This expression predicts the size of the largest drops produced by a circular water jet under specified conditions. A size distribution can then be calculated if these assumptions are made:

- The distribution is log-normal.

- The geometric standard deviation (σ_g) is similar to values for hydraulic spray nozzles.
- The maximum drop sizes observed by Ingebo are two standard deviations larger than the MMDs.

A description of the breach must be assumed as well--this would depend on the accident parameters. These equations require assuming a circular equivalent for the breach.

The relationship between the various means of a log-normal distribution (Mugele 1951) may be expressed as:

$$d_{pq} = \bar{X} \exp \left[\frac{p + q - 6}{4\delta^2} \right]$$

where

$$\bar{X} = \text{MMD}$$

$$\delta = (0.5/\ln \sigma_g)^{1/2}$$

σ_g = geometric standard deviation.

P and q are indices which define the droplet means of interest (e.g., linear, volume, Sauter, etc.). An arbitrary mean diameter, d_{pq} , is defined by

$$d_{pq} = \frac{\sum_{i=1}^{i=j} n_i d_i^p}{\sum_{i=1}^{i=j} n_i d_i^q}$$

where

n_i = number of drops in the i^{th} size group

j = number of size groups.

This enables calculation of an MMD of particle size from a breach of known diameter at specific temperatures.

Weiss (1956) concluded that atomization of liquids by large, high-velocity airstreams occurs by direct action of the airstream on the exposed liquid surfaces. He found relative velocity between liquid and airstream to be of primary importance. While physical properties of the fluids do offset spray fineness, their net influence is less critical. Geometry and operation of the injectors were considered of lesser importance, particularly at high velocities.

Empirical correlation of Weiss' experimental atomization results yielded the dimensionless equation:

$$\left(\frac{X \rho_A V^2}{\sigma_L}\right) = 0.61 \left(\frac{V \mu_L}{\sigma_L}\right)^{2/3} \left(1 + \frac{10^3 \rho_A}{\rho_L}\right) \left(\frac{W \rho_L \sigma_L A \mu}{\mu_L^4}\right)^{1/12}$$

where

X = MMD, μm

V = relative air velocity, fps

W = mass injection rate

μ_L = liquid viscosity, centipoise

ρ_A = air density, lb/ft^3

σ_L = surface tension, dyne/cm

μ_A = air viscosity, centipoise

ρ_L = liquid density, lb/ft^3 .

Most studies considered round sprays; however, one investigator looked at flat sprays. Dorman (1957) found that flat spray nozzles produced fairly large particles, and he developed a prediction equation for low viscosity liquids:

$$D_g = 12 (Q/\theta)^{\frac{1}{3}} \sigma^{\frac{1}{3}} \rho^{\frac{1}{6}} \mu^{\frac{1}{2}}$$

$$D_o = 4.4 (Q/\theta)^{\frac{1}{3}} \sigma^{\frac{1}{3}} \rho^{\frac{1}{6}} \mu^{\frac{1}{2}}$$

where

- D_g = geometric median diameter, μm
- D_o = Sauter diameter (mean-volume-surface), μm
- Q = liquid throughput, cm^3/sec
- θ = spray angle, radian
- σ = surface tension, dyne/cm
- ρ = liquid density, g/cm^3
- P = pressure, psi.

Some general observations on sprays and atomization have been made in other studies. Atomization of liquids has been summarized by Marshall (1954). The surface energy of droplets formed by jets amounts to less than 1 percent of the total energy requirements of efficient hydraulic nozzles. The energy associated with the maximum jet velocity (kinetic energy) has the same order of magnitude as the surface energy, but the largest energy component is that required to overcome viscous forces in achieving a high volumetric flow rate through a small orifice to form a small diameter jet. Breakup is characterized by violent disruption and tearing apart of the liquid, and at very high relative velocities between the gas and liquid, this disruption appears to be almost explosive.

A relationship was found between the size of drops of liquid and the slowest velocity of the airblast that would shatter them (Lane 1951). Atomization of the original drop becomes progressively finer as the pressures in the gun are increased to about 10 atm. Further pressure increase did not change the particle size beyond an MMD of 15 μm .

Merrington (1947) found the mean drop size dependent on the relative speed V of the jets to the air and on the viscosity of the liquid; the diameter was increasingly proportional to the relative velocity between jet and surrounding air.

The distribution of the liquid between the water plume and spray droplets generated should be considered, since the droplets are more likely to become airborne. An empirical correlation for the mass distribution in a water plume was developed in terms of fraction of water remaining in the water stream (Wayne 1976):

$$q/q_0 = [C (e_m - 1)^n + 1]^{-1}$$

where

q/q_0 = fraction of water remaining in the water stream

C, n = empirical constants

e_m = break-up parameters (m not defined):

$$e_m = \frac{\rho_g}{\rho_w} \left(\frac{V_g L}{V_w D_0} \right)^2 (D_0)^{0.5}$$

where

L = distance jet penetrated into gas stream, in.

D_0 = nozzle diameter, in.

V_w = water jet velocity, ft/sec

ρ_g = gas density, lb/ft³

ρ_w = water density, lb/ft³

V_s = gas stream velocity, ft/sec

for

$$e_m > 18 \quad C = 0.0000312$$

$$n = 3.83$$

$$e_m > 1 < 18 \quad C = 0.00521$$

$$n = 2.01$$

Penetration of a gas stream with a spray is another variable. It will vary with spray angle and nozzle (opening) shape. In Wayne's tests (1974), sprays from flat nozzles penetrated only 50 to 60 percent as far as solid jet nozzles.

In order for any force to exert a disruptive action sufficient to produce particles of a desired fineness, the magnitude of the force must equal or exceed any consolidating action exerted by surface tension (Lapple 1967).

Various types of forces exerted on drops are compared in Figure 4.1, which illustrates how these forces vary with drop size. Forces are expressed in terms of a pressure corresponding to a variety of conditions, as shown in Table 4.1. Lapple found that final droplet sizes are a function of Weber number and all droplets will subdivide until a critical size is reached.

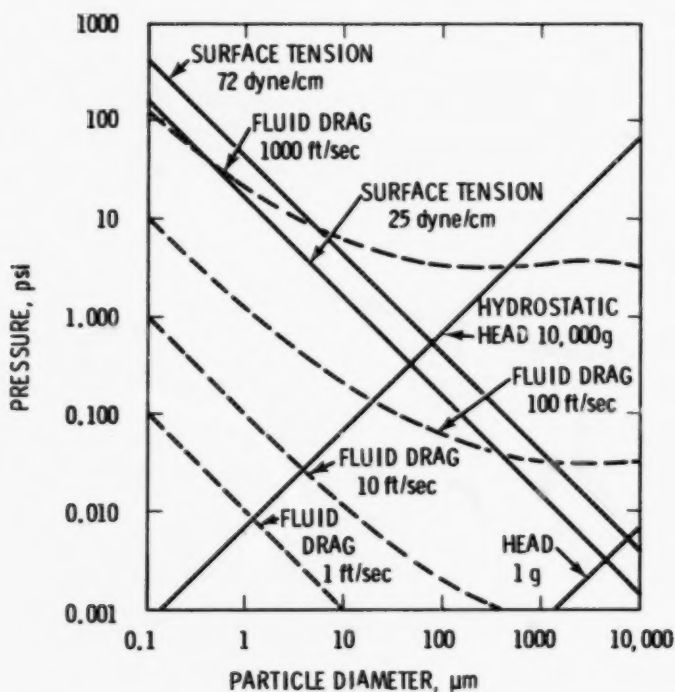


FIGURE 4.1. Pressure Exerted on Drops by Various Mechanisms

4.4 FLASHING SPRAYS

When a liquid moves isothermally from a higher pressure zone to a lower pressure zone it may cross the equilibrium pressure for liquid at that temperature and disintegrate into a spray as a result of the necessity for some of its mass to shift into the vapor state in order to maintain equilibrium. When this is the case, the liquid is said to flash (Orr 1966). Superheat of the upper zone relative to the lower zone of a system produces flashing.

TABLE 4.1. Forces of Droplets Expressed in Terms of Pressure

Mechanism or Pressure Source	Magnitude of Pressure Due to Source	Assumed Values for Plot
Surface tension	$4\sigma_j/D_p$ internal drop pressure	$\sigma_j = 25 \text{ and } 72 \text{ dynes/cm}$
Hydrostatic head (gravity or centrifugal)	$\alpha \rho_j D_p/2$ (at drop midpoint)	$\rho_j = 1 \text{ g/cm}^3$ $\alpha = \text{gravity and } 10^4 \text{ gravities}$
Fluid drag (pressure and shear)	$C_D \rho_g u_r^2/2$	$\rho_g = 0.001185 \text{ g/cm}^3$ } AIR $\mu_g = 0.0184 \text{ cp}$ } AT 25°C 1 atm
	$C_D = \psi (N_{\text{Repg}})$	$u_r = 1, 10, 100, \text{ and } 1,000 \text{ ft/sec}$
	$N_{\text{Repg}} < 0.2:$ $C_D = 24/N_{\text{Repg}}$	(slip-flow, compressibility, and droplet deformation effects neglected)
	$N_{\text{Repg}} > 1000:$ $C_D \approx 0.44$ (for spheres)	

Some general observations on particles generated by flashing sprays have been found in the literature review:

- Drops formed by a jet of superheated fluid are significantly smaller than those formed by subcooled jets.
- Homogeneous nucleation and subsequent rapid formation of vapor are the processes involved.
- No single model is able to predict drop size under spray conditions.
- Empirical data can be used for estimating drop sizes.

A jet of liquid is shattered by the rapid growth of bubbles within itself when flashing occurs. When the Weber number is greater than 12.5 for relatively low viscosity liquids, the bubble growth rate is sufficient for the jet to be shattered (Brown 1961). The mass fraction of the liquid flashed was 7.8 percent for water (at 286°F). Mishima (1976) calculated from this work

that <0.3 percent would be in the size range that could be transported downwind and constitute an inhalation hazard.

The rate at which a non-viscous, superheated liquid is jetted from a container depends on the size of the orifice, the temperature of the liquid, and the pressure on the liquid (Owstrowski 1966). He calculated the volume flow rate of drops by summing over drop-size ranges.

Estimates of drop sizes ranging from 16 to 76 μm were estimated for LOCA conditions (Koestel 1980). The best estimate was that the drop size (for local conditions) would be <16 μm due to the combined effects of heterogeneous nucleation and of aerodynamic atomization. Estimations of drop sizes produced by a source liquid approximately 315°C (600°F) are shown in Table 4.2.

Drop size from shattered sprays was found to be a function of spray temperature for a given diameter in work by Gooderum and Bushell (1969). They found no systematic variation with spray velocity or ambient pressure. These correlations can be used to predict the mean drop size produced by a superheated water jet. Figure 4.2 shows the range of normalized drop size found in these experiments as a function of temperature.

TABLE 4.2. Fragmentation Drop Size Estimates

Method of Drop Formation	Drop Size, μm
Thermal fragmentation, homogeneous nuclea- tion	16
Thermal fragmentation, heterogenous nuclea- tion	76
Aerodynamic atomiza- tion	21-35

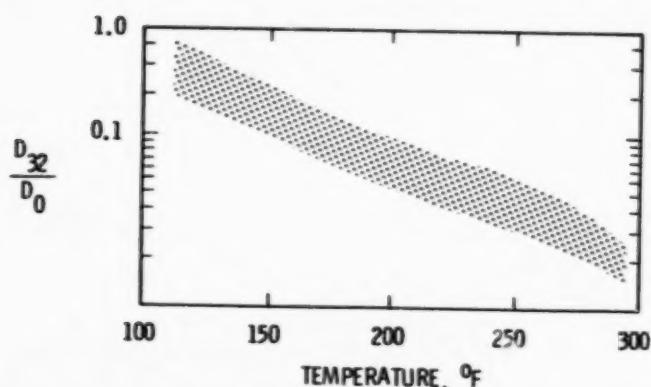


FIGURE 4.2. Normalized Mean Drop Size as a Function of Spray Temperature (centerline values)

The Hatch-Choate transformation can be used to calculate the count median diameter or MMD (Stockman 1977):

$$\log d_{gm} = \log d_{gc} + 6.908 \log^2 \sigma_g$$

where

- d_{gm} = geometric median dia by mass
- d_{gc} = geometric median dia by count
- σ_g = geometric standard deviation.

4.5 EVAPORATION

The evaporation phenomenon is complex. The bulk of a droplet evaporates immediately. The evaporation process is non-stationary and occurs in a medium with unequal temperature and vapor concentration. Drops move irregularly relative to the medium and are more or less deformed, while circulation arises within the drops (Fuchs 1959). Heat transfer occurs by conduction, convection, and radiation. The theory is very complex and will not be considered here.

Particles in air acquire and lose moisture by molecular transfer to and from the particle surface as the relative humidity of the air increases and decreases (Corn 1976). For stationary spherical water drops in air the rate of evaporation is proportional to the surface area of the drop. Experiments

with free-falling 5 to 70 μm radius drops in air at temperatures 0 to 40°C and 10 to 100 percent RH suggest quasi-stationary steady-state theory (Kinser 1951). Evaporation times of pure liquid droplets in air have been calculated and are listed in Table 4.3 (Corn 1976).

Accident generated aerosols would not be of pure water. Substances in the droplets can retard or accelerate evaporation, and so this work can only be a rough guide to droplet behavior.

Another calculation of the evaporation of falling water spray also indicated rapid evaporation of water drops falling as a water spray (Walton 1968).

The life of water drops and the distance of fall before complete evaporation of water drops of various sizes were calculated (see Table 4.4) using an equation for the rate of evaporation of a ventilated drop and allowing for changing drop size and velocity during fall. Both life and distance are inversely proportional to the difference between the vapor pressure of the drop and the partial pressure of water vapor in the air, the vapor pressure of drop being the saturation vapor pressure at the wet bulb temperature. This vapor pressure difference between drop and air when measured in mm Hg is approximately equal to one-half the wet bulb depression measured in °C.

Orr (1966) gives other examples (shown in Table 4.5) of the lifetimes of water droplets before they evaporate into air.

TABLE 4.3. Evaporation Times for Water Droplets in Still Air at 20°C and P = 760 mm Hg

Droplet Radius, μm	Evaporation Time, sec	
	0% RH	50% RH
25	7.06×10^{-1}	1.41×10^0
10	1.13×10^{-1}	2.26×10^{-1}
5	2.82×10^{-2}	5.64×10^{-2}
1	1.13×10^{-3}	2.26×10^{-3}
0.5	2.82×10^{-4}	5.64×10^{-4}
0.1	1.13×10^{-5}	2.26×10^{-5}

TABLE 4.4. Life and Distance of Fall Before Complete Evaporation of Water Drops in Air for Wet Bulb Depression of 10°C (5 mm Hg vapor pressure difference)(a)

Drop Diameter, μm	Life, sec	Distance of Fall, cm
10	0.10	0.014
20	0.38	0.23
40	1.5	3.4
100	8.3	102
200	27	1,040
400	82	7,400
1000	230	72,000

(a) for other humidities, life and distance vary inversely as the wet bulb depression.

Further information on evaporation is found in studies of evaporators. Entrainment of liquid drops follows the bursting of steam bubbles from a boiling surface. Only the smaller particles are likely to become entrained.

The maximum height attained by a drop from a boiling surface has been calculated and is shown in Figure 4.3 for different vapor mass velocities (Yamamoto 1968). These values are based on a surface tension of 62.2 dyne/cm

TABLE 4.5. Life of Water Droplets in Air

Initial Droplet Diameter, μm	Time (sec) for Complete Evaporation Under Temperature and Relative Humidity of:	
	20°C, 80% RH	30°C, 50% RH
80	33.7	9.46
60	19.2	5.37
40	8.73	2.43
20	2.35	0.64
10	0.64	0.16

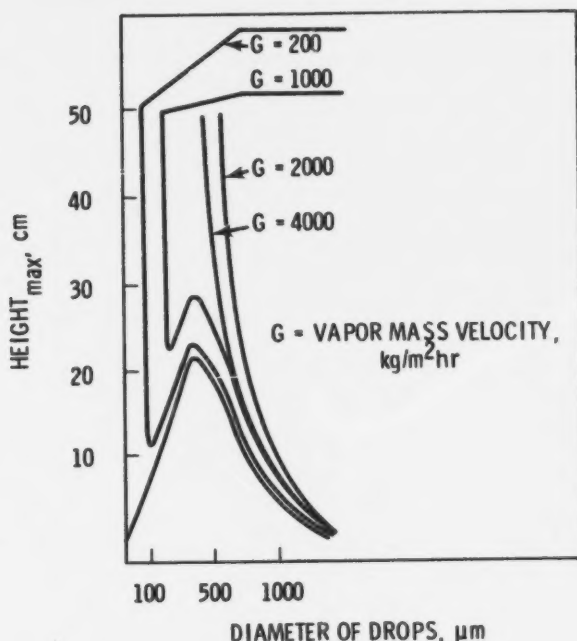


FIGURE 4.3. Calculated h_{max} in Evaporator

and specific gravity of 1.03 g/cm^3 . Experiments under normal boiling conditions (mass velocities below $400 \text{ kg/m}^2/\text{hr}$) show that height of drop above the liquid level does not vary greatly due to vapor velocity. Small drops $<100 \mu m$ are more likely to go higher and be entrained at all velocities. Chamber height and diameter will also influence liquid entrainment.

4.6 LIQUID POOL RELEASE

In an experimental study (Aubert 1974) bacteria injected into water were found in aerosols released from a basin by air moving over the water. The bacteria could be considered tracers and resuspension rates could be calculated. The number airborne varied with time and distance from the basin; the release from fresh water might be assumed appropriate in estimating releases from a pool resulting from a spill. Resuspension rates covered a range of values, with the amount airborne decreasing rapidly with increasing distances from the source pool. Based on the values calculated, $1 \times 10^{-10}/\text{sec}$ might

be appropriate for a pool release. This would assume uniform distribution in the source and airborne material.

The mass exchange between liquid and vapor is determined by vapor pressure and the pressure P or ΔP (Hickman 1965). It is believed to occur spontaneously according to the equations:

$$W_{g/\text{sec}/m^{-2}} = 583 \left(\Delta P \sqrt{\frac{M}{T}} \right) E$$

and

$$W = 583(P_S - P_G) \sqrt{\frac{M}{T}} \times E$$

where

ΔP = change in pressure, mm Hg

T = temperature, °C

M = not defined (probably molecular weight)

P_S = surface vapor pressure

P_G = bulk gas vapor pressure

E = correction coefficient.

The varying quantity W , characteristic of any substance and temperature, deserves unit status and could be called the Knudsen or Knud (Kn) in $g/m^2/\text{sec}^1$. The coefficient E is included to allow for the failure of measurement to agree with theory; that is,

$$E = \frac{W_{\text{obs}}}{W_{\text{calc}}}$$

The value E is always less than unity, a fact that is attributed either to measurement errors or to a real failure of molecules to cross and recross the interface in the calculated numbers. Experimental values of E ranged from 0.036 to 0.52 for water at 25°C. It would appear that these formulations could be used to estimate pool releases.

4.7 SPARGING

Particles are entrained in gas and vapor bubbles and are carried to the atmosphere as bubbles break through a pool surface in a process known as sparging. Literature on sparging could be used to estimate releases from boiling pools or pools with gas bubbles rising through the pool.

The formation of droplets through collapse of a bubble dome is considered unavoidable (Perry 1973). Droplet sizes are generally $<25\text{ }\mu\text{m}$, which means that their terminal velocities are low and they will be entrained. Because of their small size they contribute little on a weight basis ($<0.001\text{ lb liquid/lb vapor}$) although they dominate in sheer numbers. Rupture of bubbles leaving a liquid phase in the separator produced droplets $<40\text{ }\mu\text{m}$.

There is a dependence on sparge velocity to E, the entrainment ratio: (lb entrained liquid/lb vapor) (Perry 1973). In the low regime, it is proportional to velocity; above $E < 0.0001$ it will vary roughly with the velocity.

In an experimental study (Pueschel 1974) a particle production rate (from a bubbler) by sparging of sea water was $1.7 \times 10^{-9}\text{ g/cm}^2\text{/sec}$ from the experimental surface, with a $0.1\text{ }\mu\text{m}$ MMD particle size. In these studies, both vapor and particles became airborne through sparging.

Seawater experiments also indicate that aerosols produced by sparging can contain enhanced levels (relative to Na) of contaminants depending on factors such as rise distance (Van Grieken 1974). These fractionation effects may exceed a factor of 10 and appear to be the rule rather than exception. Van Grieken concluded it was unwise to assume the composition of a marine aerosol to be the same as sea water.

The very small inertia of the gas in a bubble permits internal circulation to die away rapidly (Orr 1966). For bubbles that are less than about 2.5 mm dia the continuous-phase mass-transfer coefficient is related by an expression similar to previous relationships:

$$Sh = 2.0 + 0.31(Ra)^{0.33}$$

where

Sh = Sherwood number, mass transfer coefficient

Ra = Rayleigh number;

$$Ra = \frac{(d^3 \Delta \rho g)}{(\mu_L D_{fL})}$$

where

d = bubble diameter

$\Delta \rho$ = difference in gas and liquid densities

g = acceleration of gravity

μ_L = liquid viscosity

D_{fL} = diffusion coefficient in the liquid phase.

The correlation makes no allowance for velocity. This means that it gives a minimum coefficient and may be expected to hold best for mild sparging and agitation conditions where the bubbles behave as rigid noncirculating spheres.

Huang (1978) cited tests on the release of PuO_2 during heating of a test sample from 25° to 560°C, which indicated a release due to sparging of entrapped gases. Decontamination factors obtained ranged from about 12 to 73.

4.8 SUMMARY

No direct method of estimating airborne spill releases was found, e.g., weight percent airborne from a source volume or mass. Estimates of airborne concentrations are available and have been used to evaluate the aerosol generated, although there are large uncertainties with this method.

There appears to be considerable information available to estimate well-defined spray releases, but only a portion has been included in this report. Perry's text is a standard reference that can be consulted for equations representing releases in specific spray situations. The flow rate from a round opening (e.g., nozzle) can be used to estimate the total liquid discharged. This requires knowledge of the opening size, discharge coefficient, static pressure losses, and pressure differential.

While spray releases of liquids seem fairly well defined, accident stresses will impose factors such as impaction, satellite formation, secondary breakup, etc. These must be described before an estimate of total airborne mass and size is made. Some of these methods have been included in this chapter.

Evaporation poses the question of whether the droplet is a pure compound or contains a solute that remains airborne after evaporation. It would appear that any contamination associated with the evaporating solution would subsequently become airborne.

Finally, sparging can produce an aerosol with large numbers of fine particles.

5.0 CONDENSATION

Condensation aerosols are formed when supersaturated vapor condenses or when gas-phase reactions result in a solid reaction product, such as soot (Fuchs 1964). They might be anticipated after an accident that generates aerosols. Particles in air acquire and lose moisture by molecular transfer to and from the particle surface as the relative humidity of the air increases and decreases (Stern 1976). In condensation aerosols solid particles are often loose aggregates of a very large number of primary particles of a regular crystalline or spherical form (Fuchs 1964).

Condensation aerosols are entirely different from dispersion aerosols. The particle sizes of condensation aerosols are usually much finer (0.1 to 30 μm). Condensation aerosols with a solid disperse phase are called smokes; those with a liquid disperse phase are called mists or fogs (Perry 1973). In order to form a fog, nuclei must first be found. After an accident, foreign nuclei would be abundant and would provide opportunity for nucleation.

Industrial fogs show a broad size distribution. For example, the particle size distribution in an absorption tower of a contact H_2SO_4 plant was 0.75 μm MMD (σ_g^2). The sizes found are smaller than those found for entrainment, atomization, and even for high speed droplet ruptures. The mist loading was 0.73 and 0.62 mg/l.

Condensation could be assumed to be more of a continuous than instantaneous source term. It has been studied in pipes (Lapple 1956), where the pressure drop of a condensing vapor can be calculated using traditional methods. Condensation in pipes is complicated by the changing properties of the mixture due to constantly changing pressure, temperature and fraction condensed. This would also be true of accident-generated condensation aerosols.

5.1 ATMOSPHERIC CONDENSATION AEROSOLS

Atmospheric condensation aerosols were studied by Perrie (1950). These studies help us to understand the aerosols that result from accident stresses. Nuclei are necessary for the formation of condensation aerosols without

enormous supersaturation. These nuclei may or may not be hygroscopic (having an ability to accelerate condensation). The nonhygroscopic particles, e.g., coal or floor dust, do not appear to be particularly satisfactory as condensation nuclei. Larger dust particles can be mechanically picked up by falling precipitation. Condensation occurs more readily on larger droplet nuclei than on smaller ones.

Vapor pressure over a convex surface, such as a drop, is greater than that over a plane surface of the same liquid at the same temperature. It has been suggested that due to this phenomenon nonhygroscopic porous surfaces are satisfactory nuclei when moisture is in the pores.

The electric charge carried by a droplet nucleus has been suggested as one of the attracting forces causing condensation. However, it has been shown experimentally that the charges on nuclei seldom exceed one electron charge. A charge of many times this magnitude would have only an infinitesimal effect on the hygroscopic properties of the nucleus. It seems very doubtful if this effect is of any great importance.

5.2 CONDENSING SYSTEMS

The behavior of condensing systems is determined by the interaction of thermodynamic and rate processes (Friedlander 1977), some of which are listed below.

- Adiabatic Expansion. Reversible adiabatic expansion can lead to a decrease in both temperature and pressure of an initially unsaturated gas up to the point of condensation.
- Mixing. Condensation by mixing can result when a hot gas carrying a condensible vapor is mixed with cool gas.

Vapors generally must be greatly supersaturated (4.2 times the saturation level of water vapor) before condensation will occur in the absence of foreign particles (Magill 1956); if particles are present to act as condensation nuclei, little supersaturation is necessary. The following condensation nuclei, ranging in size from about 0.001 to 0.1 μm , would be abundant in an accident scenario:

- inert particles possessing plane or porous surfaces or carrying an electrical charge
- substances that have a strong chemical affinity for the vapor.

5.3 SPRAY CONDENSATION

Pasedag (1971) modeled condensation and coalescence of spray droplets. The results show that the reduction in the mass transfer surface area would be about 10 percent for the spray system. A spectrum of drop sizes is produced by spray nozzles, and Pasedag has developed models to estimate changes in this initial distribution due to condensation and coalescence.

5.4 STUDIES OF CORE DISRUPTIVE ACCIDENTS

Mechanisms for particle generation that determine the quantity and range of particle sizes have been studied as related to nuclear core disruptive reactor accidents (Kennedy 1977a). The range of particle sizes observed in tests at ORNL were reproduced by a model developed between the fuel vapor and argon. The particles were generated by homogeneous nucleation condensation, and a detailed formation and growth model was developed.

Two categories of condensation nuclei are assumed: 1) those within the bulk vapor phase and 2) those in the thermal boundary layer adjacent to a physical barrier.

5.4.1 Nuclei Formation--Category 1

Concern with supersaturation in a vapor stream led to development of a model predicting homogeneous nucleation condensation of metal vapors expanding through the converging section of a nozzle (Hill 1973). Classical nucleation growth theories were used.

Storebo (1974) developed a model to calculate three processes involved in the evolution of particle-size distributions formed by condensation of material in a nuclear explosion fireball. These processes are 1) nucleation of particles directly from vapor, 2) condensation on particles brought in with the air and 3) agglomeration of particles in the fireball.

5.4.2 Nuclei Formation--Category 2

Fog that is formed from homogeneous nucleation condensation in the thermal boundary layer increases the rate of diffusion-limited vaporization from heated surfaces (Rosner 1968). Homogeneous nucleation begins when a critical level of supersaturation is reached locally within the boundary layer. Looking at heterogeneous condensation on cold surfaces, Rosner and Epstein (1970) developed a criterion for predicting the onset of fog formation due to homogeneous nucleation condensation. Later they developed a more detailed homogeneous nucleation model.

5.5 OTHER CONDENSATION MODELS

Ohmburg and Olander (1971) developed a theoretical model describing details of the condensation processes.

A model predicting the critical supersaturation ratio (ratio of vapor pressure to saturation pressure) for the inception of homogeneous nucleation condensation in a diffusion cloud chamber was developed by Katz and Ostermier (1967).

Epstein and Cho (1975) presented the results of calculations which indicated hot vapor may condense into an aerosol (fog) within the thermal boundary layer near any available cooler surface.

Kennedy et al. (1977b) assumed homogeneous nucleation provided the initial source and made a computer simulation of events. Vaporized UO_2 , quenched as it mixes with cold argon gas, produces increasingly supersaturated vapor states and causes particles to nucleate directly from the vapor. Once nucleated particles grow, the remaining vapor condenses on them. An approximate log-normal particle size distribution function results. The median particle size was very small: $3.0 \times 10^{-2} \mu\text{m}$ radius. Ninety-nine percent had a radius less than $2.5 \times 10^{-2} \mu\text{m}$, with the maximum radius $7.5 \times 10^{-2} \mu\text{m}$. This compared well with experimental work.

Kennedy (1977a) made the following general conclusions:

- The condensation model based on homogeneous nucleation that he developed could reproduce experimentally-observed particle size distributions.

- This model had to assume rapid cooling of the UO_2 vapor resulting from rapid mixing with argon gas
- The model did not reproduce experimentally-observed bimodal distribution.

The total number of particles formed is a function of the total mass of vapor, although the distribution of particle sizes is not.

5.6 GAS-TO-PARTICLE CONVERSION

Reactions of gas-to-particle conversions result in an increase in the airborne mass concentration, with the mechanisms and rate of conversion to particulate matter of interest. Two general types of conversion (Friedlander 1977) are caused by:

- gas phase processes, where a supersaturated state is produced which then collapses due to aerosol formation
- particulate phase processes, where molecules from the gas may react on the particle droplet or in a droplet.

Table 5.1 summarizes gas-to-particle conversion mechanisms. Calculations are available to estimate growth and are summarized in Table 5.2.

5.7 SUMMARY

A large body of information has been generated relating to LOCAs for reactors. This information could be used to evaluate postulated condensation aerosols of other nuclear fuel cycles. A limited discussion has been included here.

TABLE 5.1. Examples of Gas-to-Particle Conversion Mechanisms

Homogeneous Nucleation

Physical Processes Producing Supersaturation

- adiabatic expansion
- mixing
- conductive cooling
- radiative cooling

Gas Phase Chemical Reaction

- single condensable species (classical theory)
- multicomponent condensation (heteromolecular theory)

Heterogeneous Condensation

Transport-Limited Chemical Reaction

- diffusion, $d_p \gg 1 \text{ cm}$
- molecular bombardment, $d_p \ll 1 \text{ cm}$

Surface-Controlled Chemical Reaction

Particulate Phase-Controlled Chemical Reaction

TABLE 5.2. Growth Laws for Gas-to-Particle Conversion

<u>Mechanism</u>	<u>Growth Law, dv/dt</u>
Diffusion ($d_p \gg 1$)	$\frac{2\pi D d_p v_m (p_1 - p_d)}{kT}$
Molecular bombardment ($d_p \ll 1$)	$\frac{\alpha \pi d_p^2 v_m (p_1 - p_d)}{(2\pi m k T)^{1/2}}$
Surface reaction (all sizes)	$\frac{\alpha \pi d_p^2 v_m p_1}{(2\pi m k T)^{1/2}} (\alpha \ll 1)$
Droplet phase reaction	$\frac{\pi d_p^3}{6\rho_p} (\sum v_i) r$

where

D = diffusion coefficient, cm^2/sec

d_p = particle diameter, cm

v_m = condensing species molecular volume, cm^3

TABLE 5.2. (contd)

v	= particle volume, cm^3
p_1	= pressure, mm Hg
p_d	= equilibrium vapor pressure, mm Hg
k	= mass transfer coefficient, cm/sec
T	= absolute temperature, $^{\circ}\text{K}$
α	= 1, coefficient for rigid elastic spheres
m	= mass of particle or molecule, g
ρ_p	= particle density, g/cm^3
M_i	= molecular weight
ν_i	= kinematic viscosity
r	= radial coordinate, cm

6.0 AIRBORNE CHEMICAL REACTIONS AND CONVERSIONS

Gases can react with condensation nuclei and other small particles in the air during transport. After reaction, the gas loses its identity and is transported as the host particle with which the gas initially reacted. This type of reaction would undoubtedly be a component of many accidents; however, information on these reactions as they relate to accidents could not be found in the literature. Some work with atmospheric aerosols is therefore reviewed to illustrate the components involved.

The substances reacting in atmospheric situations fall into two groups: natural constituents and contaminants (Magill 1956). After an accident, the contaminants could be assumed to be in high concentrations, calling for careful interpretation of the kinetic and photochemical data in the literature.

Atmospheric chemical reactions in air pollution have been classified (Butt 1971) as follows:

- photochemical: $\text{NO}_2 + h\nu \rightarrow \text{NO} + \text{O}$
- ozone: O_3
- nitrogen oxide family: NO_x
- free radical reactions: $\dot{\text{R}}, \text{RO}, \text{ROO}, \text{PAN}$
- other: $\text{SO}_2, \text{H}_2\text{S}, \text{N}_2\text{O}$

6.1 HETEROGENEOUS PROCESSES

Heterogeneous processes can involve particles and the generation or reaction of gaseous pollutants. Judeikis (1973) assessed atmospheric reactions involving CO, NO, SO_2 , olefins and NO_2 . He observed that a large number of reactions can occur even in the simplest systems. A well known example of a heterogeneous reaction involving aerosols is the oxidation of sulfur dioxide in water droplets.

Judeikis cited a collision model for particle-gas interaction that relates the frequency of collision K_c between gaseous pollutant molecules G , and particulate matter with total surface A :

$$K_c[G]A = \left(\frac{RT}{2\pi M_g}\right)^{1/2} [G]A$$

where

[G] = concentration

M_g = molecular weight

R = gas constant

T = absolute temperature.

Only a fraction of the gas-particle collisions (ϕ) will lead to removal of G from the gas-phase; the rate of disappearance of G can be written:

$$\frac{-d[G]}{dt} = \phi K_c[G]A = \phi \left(\frac{RT}{2\pi M_g}\right)^{1/2} [G]A = R'_{calc}[G]$$

If M_g is in kg, $K_{calc} = 0.0272 \sqrt{\frac{g}{M_g}} \text{ s}^{-1}$

Judeikis used experimental data and calculated values of ϕ required for heterogeneous processes to compete with the homogeneous reactions. These values are shown in Table 6.1.

TABLE 6.1. Values of k'_{expt} and ϕ for Selected Reactions

Molecule	CO	NO	SO ₂	Olefins	NO ₂
Reaction	(a)	(a)	ϵ	(a)	$\neq$
Reactant	OH	O ₃	O ₂	O ₃	Light
$k'_{expt}, \text{ s}^{-1}$	2.4×10^{-7}	4.1×10^{-2}	1 to 20×10^{-7}	1 to 100×10^{-5}	5.8×10^{-3}
ϕ s	1.5×10^{-6}	2.6×10^{-1}	1 to 20×10^{-6}	0.6×10^{-4} (C ₂ H ₄) 1 to 100×10^{-4} (C ₆ H ₆)	4.6×10^{-2}

(a) Oxidation: ϵ = photooxidation; $\neq$ = photodecomposition; ϕ s = fraction of collisions required for reaction (or adsorption) to give $k'_{calc} = k'_{expt}$. Photooxidation of SO₂ and photodecomposition of NO₂ would be unimportant during evening hours.

This data indicates that a value of $\phi = 0.26$ would be required for the rate of heterogeneous oxidation of NO to equal that of a gas phase reaction. Under certain favorable conditions surface-catalyzed reactions in polluted atmospheres can compete with and, in some cases, dominate homogeneous gas-phase reactions.

6.2 ADSORPTION

As Liberti (1970) considered surface mechanisms, he observed that particulates in an unstable system are in continuous evolution. Particulates behave as an adsorption medium having two physicochemical properties: 1) they act as a carrier for a variety of pollutants which are concentrated on the surface, and 2) they act as catalysts for various reactions. Metallic components will influence the reactions of particles, and organics can also be associated with the particles.

Judeikis (1973) had noted that information on the catalytic activity of generated particles was unavailable.

Finely divided carbon (soot) particles can play a major role in the catalytic oxidation of surface dioxide to sulfate in polluted atmosphere (Novakov 1974).

6.3 AEROSOL SURFACE AREA

McMurray and Friedlander (1978) related aerosol surface area to rate-of gas-to-particle conversion. For monodisperse aerosols and aerosols whose size distributions are self-preserving, the surface area per unit volume of gas can be expressed as a function of the rate of gas-to-particle conversion dv/dt and time t . Chemical reactions in the gas phase may result in the formation of condensable particles. Molecules can either deposit on pre-existing particles or form new particles by collisions among themselves. The area surface area per unit volume of gas should be essentially independent of the chemical nature of the condensing species.

Size distributions of aerosols growing by gas-to-particle conversion probably become self-preserving after the surface area stops changing. The

self-preserving surface area A is a weak function of time, and increases as a function of the rate of aerosol formation. Experimental and theoretical aerosol surface areas for a given rate of aerosol formation are compared in Figure 6.1. A simplified equation to predict the surface area, assuming time to be 1 hr, density 1.46 g/cm^3 , and temperature 300°K is:

$$A = 6.23 \times 10^3 F^{3/5}$$

where

A = aerosol surface area cm^2/cm^3 air

F = cm^3 aerosol/ cm^3 air/sec.

For the special case of constant dv/dt , surface area varies as $(dv/dt)^{3/5} t^{1/5}$, and is essentially independent of the nature of the secondary aerosol.

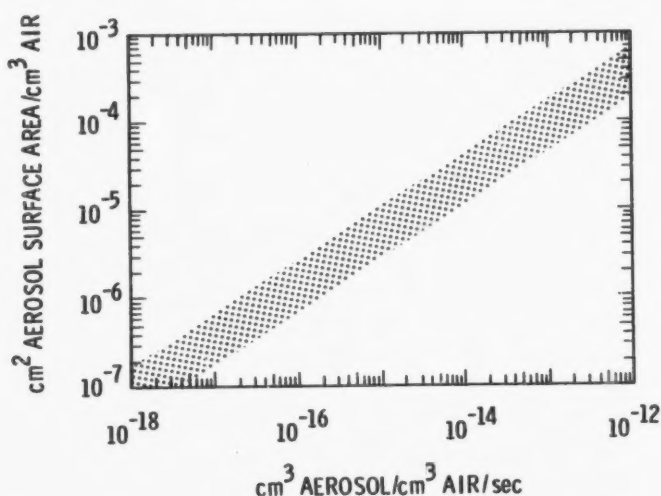


FIGURE 6.1. Comparison of Experimental Aerosol Surface Areas for a Given Rate of Aerosol Formation (range of values shaded)

6.4 SUMMARY

For the most part, the authors cited were primarily interested in atmospheric (outdoor) pollution, e.g., smog. The fuel cycle accidents under consideration are indoor, and this could change the importance of contributing parameters. For example, photochemical reactions decrease in importance. Interior surfaces could enhance the probability of catalyzed reactions due to either additional surface area for adsorption or reactions taking place, or by being closer to the pollutant source where higher concentrations of the particles exist.

More information is becoming available on post-accident chemical reactions of cesium and iodine; a review of this topic is beyond the scope of this report, however.

Post-accident reactions could lead to the formation of objectionable products, or objectionable products may be converted into harmless ones, and research has been carried out to explore these mechanisms (atmospheric reactions) in detail. However, at this point in our study, a better identification of the accident is required before much speculation can be made on the nature of chemical reactions.

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